

Nonlinear processes in two-photon pumped
atomic vapors

Dissertation der Fakultät für Physik,
Ludwig-Maximilians-Universität
München

vorgelegt von
Angela M. Guzmán de García
aus Bogotá, Kolumbien.

München, 18. Juni 1984

Nonlinear processes in two-photon pumped
atomic vapors

Dissertation der Fakultät für Physik,

Ludwig-Maximilians-Universität

München

vorgelegt von

Angela M. Guzmán de García

aus Bogotá, Kolumbien.

München, 18. Juni 1984

1. Gutachter: Dr. habil. P. Meystre

2. Gutachter: Prof. Dr. H. Wolter

Tag der mündlichen Prüfung: 27. September 1984

Gedruckt mit Unterstützung des Deutschen Akademischen Austauschdienstes.

Contents

Chapter I. Introduction

- 1.1. Statement of the problem.
- 1.2. Previous results. - Upper limits bounding and asymptotic properties concerning.
- 1.3. Previous results. - General Remarks concerning the proof.
- 1.4. Scope of the thesis.

Chapter II. The finite field axioms

- 2.1. Introduction.
- 2.2. Definition of finite fields.
- 2.3. The finite fields having p^n elements.
- 2.4. Uniqueness of the finite fields.

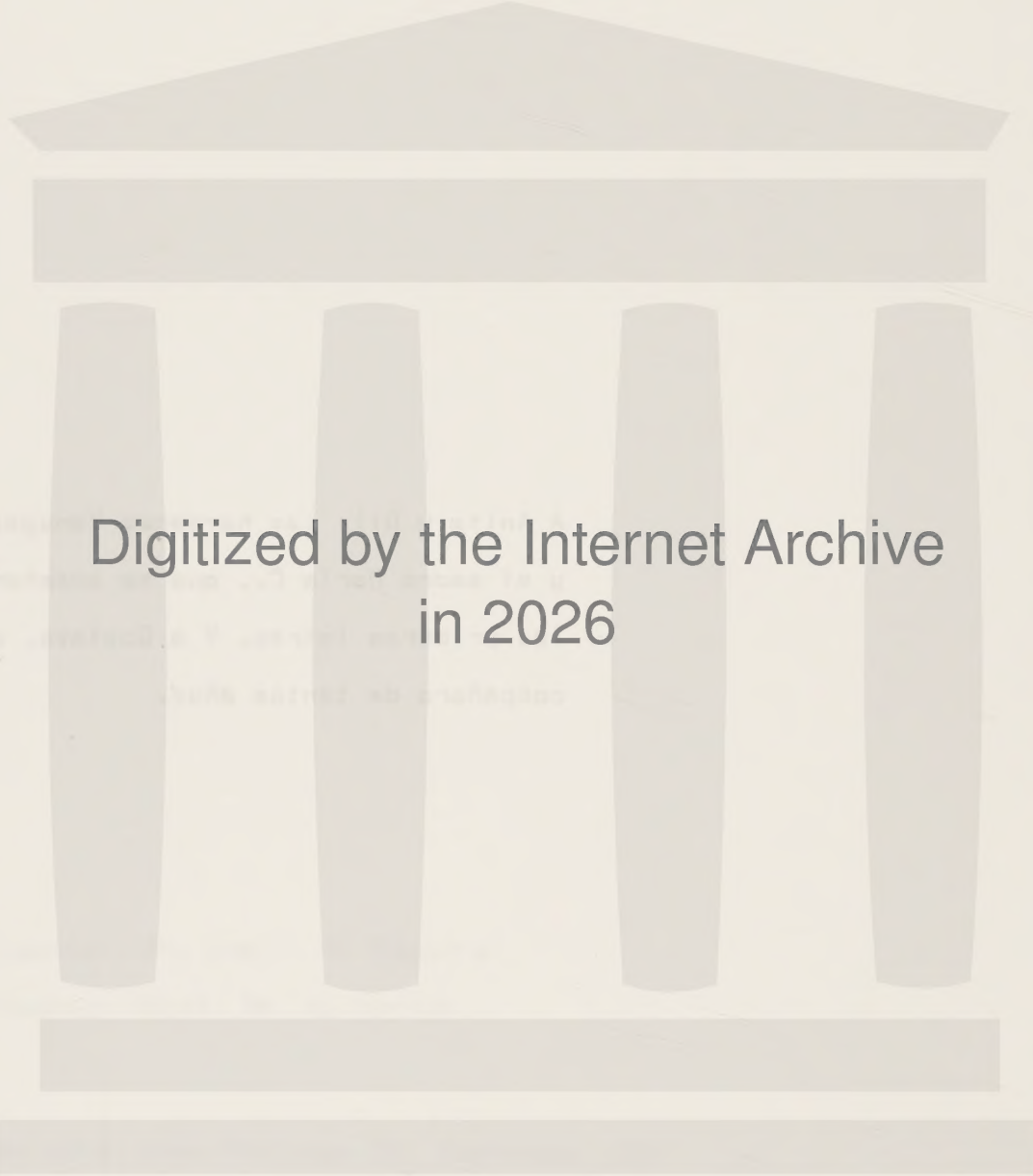
A Anita y Oli, las hermanas Vanegas,
y mi madre María C., que me enseñaron
las primeras letras. Y a Gustavo, el
compañero de tantos años.

Chapter III. Finite field axioms

- 3.1. Finite field axioms as an axiom system.
- 3.2. Field axioms.
- 3.3. Linear spaces over $\mathbb{F}_q$.
- 3.4. Extensions.
- 3.5.1. Construction of the finite field.
- 3.5.2. Generation of the finite field.
- 3.6. Connections with Galois theory.

Chapter IV. Isomorphisms and automorphisms

- 4.1. Introduction.
- 4.2. The Frobenius automorphism.
- 4.3. Results on the automorphisms of the finite field $\mathbb{F}_q$.



Digitized by the Internet Archive
in 2026

https://archive.org/details/IA41555209_0022

CONTENTS

Chapter I.	<u>Introduction</u>	
1.1.	Statement of the problem.	1
1.2.	Previous results. - Hyper-Raman scattering and four-photon parametric conversion.	4
1.3.	Previous results. - Second Harmonic Generation in gases.	7
1.4.	Scope of the thesis.	9
Chapter II.	<u>The matter-field equations</u>	11
2.1.	Introduction.	11
2.2.	Density matrix equations.	14
2.3.	The Slowly Varying Envelope Approximation (SVEA).	20
2.4.	Discussion of the method.	25
Chapter III.	<u>Steady state regime.</u>	28
3.1.	Steady-state solution in the linear regime.	29
3.3.	Field propagation.	31
3.4.	Linear Stokes gain.	37
3.5.	Saturation.	44
	3.5.1. Saturation of the Stokes field.	44
	3.5.2. Saturation of the idler field.	48
3.6.	Comparison with Cadmium experiments.	50
Chapter IV.	<u>Transient regime - Square pump pulses.</u>	53
4.1.	Introduction.	53
4.2.	The thin sample approximation.	56
4.3.	Density matrix equations in the weak probe limit.	58

4.4.	Optical nutation.	59
4.4.1.	Pumping at the Stark shifted resonance $\Delta_1' = 0$.	60
4.4.2.	Perturbative approach in decay constants.	66
4.5.	Free induction decay.	70
4.6.	Conclusions.	73
Chapter V.	<u>Gaussian pump pulses and pulse propagation.</u>	75
5.1.	Introduction.	75
5.2.	Probe gain for a Gaussian pump pulse.	76
5.2.1.	Slow decay rates.	77
5.2.2.	Fast decay rates.	82
5.3.	Pulse propagation.	84
Chapter VI.	<u>On Second Harmonic Generation in atomic vapors.</u>	91
6.1.	Introduction.	91
6.2.	Comparison with experimental findings with the available theoretical models.	93
6.3.	Why should collisions be considered?	96
6.4.	Two-photon pumping in the presence of a dc-field or l-mixing collisions.	98
6.5.	Photoelectric fields.	103
6.5.1.	Two-photon pumping.	104
6.5.2.	Phase-mismatch factor.	107
6.5.3.	DC-field mixing factor.	108
6.6.	Collisional mixing.	109
6.6.1.	A crude collisional model.	112
6.7.	Comparison with the experiment.	115
Chapter VII.	<u>Conclusions and open questions for future research.</u>	119

Appendix I. Derivation of equations for the resonant density matrix elements.	128
References.	135
Acknowledgements.	140
Vita.	141

CHAPTER I

INTRODUCTION

1.1 Statement of the problem.

Nonlinear optics owes its existence to the intense, coherent light sources provided by lasers. Its development has run parallel to that of the laser, and it has lead to a significant extension of the spectral range covered by coherent sources. The first nonlinear optical phenomenon was observed by Franken et al. /1/, who generated the second harmonic of a ruby laser by focusing it onto a quartz crystal. In later work, the phase velocity mismatch between fundamental and second harmonic was partially circumvented by the use of birefringent media, leading to longer coherence lengths and higher conversion efficiencies /2/. A substantial effort concentrated thereafter on the search for new nonlinear crystals and further improvement of second harmonic generation (SHG) conversion efficiencies. Curiously it was not until about a decade later that the study of nonlinear effects in gases received much attention. Some reasons for this delayed development are the following: first, even-order nonlinear effects (including SHG) are forbidden in centrosymmetric media; second, active atomic densities are much smaller in gases than in crystals; and third, phase mismatch reduces strongly the coherence length, and unlike in crystals, there is no medium anisotropy available to eliminate this problem. New and Ward /3, 4/ were the first to generate the third harmonic (third order nonlinear process) of a ruby laser beam focused into a gas cell. Because of phase mismatch and nonresonant pumping the conversion efficiency was however very low. A significant step, leading to orders of magnitude improvement in

efficiencies, was obtained by Harris and Miles /5-7/, who used a metal-buffer gas mixture at appropriate pressure ratio to achieve phase matching between fundamental and third harmonic. Thereafter, gases became of great interest as sources of coherent ultraviolet and vacuum ultraviolet radiation. They offer good transparency in the infrared as well as in the ultraviolet, where crystals are highly absorptive; they do not suffer irreversible damage at high intensities, and vapors of good optical quality over large dimensions are easier to prepare than crystals. For a detailed review of nonlinear processes in atomic and molecular gases, see the book by Hanna et al. /8/.

In this thesis, we investigate three nonlinear processes in three-level systems : Stimulated Hyper-Raman Scattering (SHRS), four-photon parametric conversion, and Second Harmonic Generation (SHG) in gases. For the sake of clarity we have sketched the different processes in Fig. 1.1. The relevant atomic levels are labeled $|1\rangle$, $|2\rangle$, $|3\rangle$. The states $|1\rangle$ and $|2\rangle$ have the same parity, opposed to that of $|3\rangle$; therefore, electric dipole transitions are allowed between levels $|1\rangle$ and $|3\rangle$, and between levels $|2\rangle$ and $|3\rangle$, but not between levels $|1\rangle$ and $|2\rangle$. SHRS is defined as a three-photon process, where the annihilation of two pump photons and the emission of a single Stokes photon take place simultaneously (Fig. 1.1a) . The atom undergoes the transition $|1\rangle \rightarrow |3\rangle$ in one step, by the simultaneous absorption of two pump photons of frequency ν_1 and the emission of one Stokes photon of frequency $\nu_2 = 2\nu_1 - \omega_{31}$, where $\hbar\omega_{31}$ is the energy difference between levels $|3\rangle$ and $|1\rangle$, and ω_{31} the corresponding transition frequency (*). This three-photon process is analogous to the two-photon process of Stimulated Electronic Raman Scattering (SERS). When the pump is tuned

(*) We use ν , ω to denote field frequencies and atomic frequencies, respectively. Both are angular frequencies.

close to the two-photon resonance, significant population is transferred to the upper state $|2\rangle$, and lasing to the empty state $|3\rangle$ can take place alongside the resonantly enhanced SHRS. The nomenclature in the literature is mixed, and measurements include the contribution of population-type and Raman-type processes.

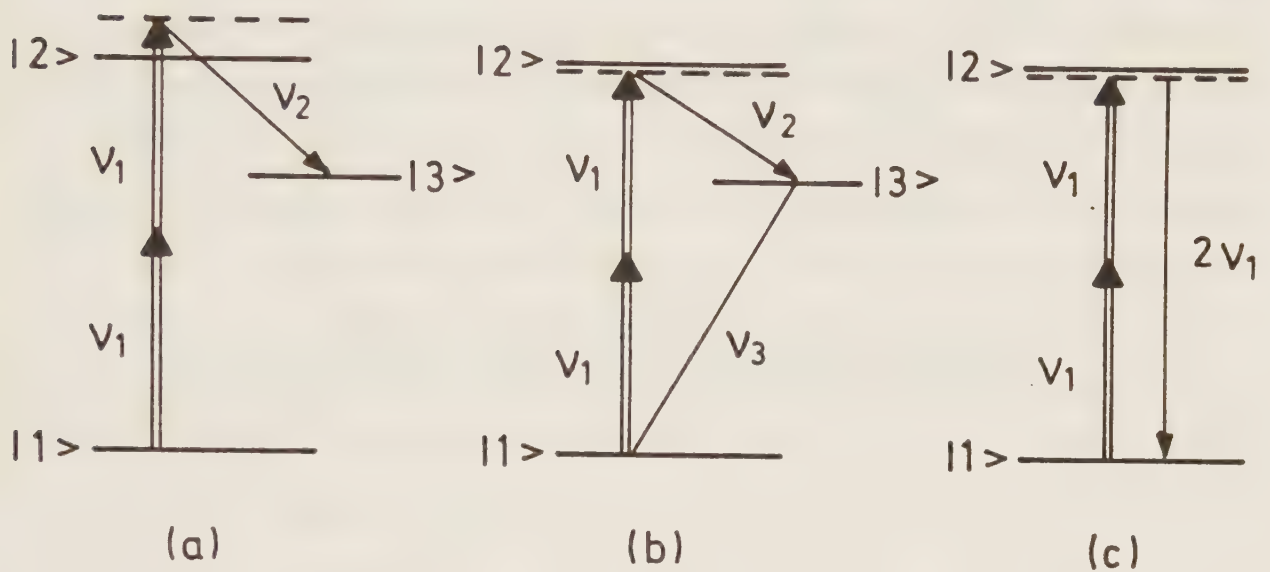


Fig. 1.1

The three nonlinear processes investigated here: (a) Stimulated hyper-Raman Scattering (SHRS). (b) Four-photon parametric conversion. (c) Second Harmonic Generation (SHG).

The four-wave parametric mixing we are interested in is shown in Fig. 1.1b: two pump photons are annihilated, and simultaneously two photons whose frequencies ν_2 and ν_3 add up to $2\nu_1$ are generated. The field at frequency ν_2 will be called here the Stokes or signal field, and that at frequency ν_3 the idler field.

Finally we will consider the second harmonic generation process (Fig. 1.1c), where two pump photons are absorbed and simultaneously one photon at twice the pump frequency is emitted. Emission at $2\nu_1$ may require mixed parity.

1.2 Previous results. - Hyper-Raman scattering and four-photon parametric conversion.

The processes described above have already been studied, both experimentally and theoretically. SHRS has been investigated in some detail /9-14,49-52/ as one of the possibilities to generate tunable infrared radiation in gases. The measurements have been carried out by taking advantage of the resonant enhancement obtained by tuning the pump laser in the region of the two-photon resonance. In this way, photon conversion efficiencies up to two percent have been attained for example in sodium vapor /10/. Most of the theoretical calculations of the nonlinear polarization (see e.g. /50/) have been based on the fifth order susceptibility $\chi^{(5)}(-\nu_2; \nu_1, \nu_1, -\nu_1, -\nu_1)$. This approximation remains valid in the low intensity pump regime, but breaks down whenever ground state depletion arises.

More complex, but also more accurate in the high intensity regime, are recent theoretical treatments /13-14,51-52/, where the density matrix formalism has been applied to investigate the dynamics of the SHRS in media consisting of four-level atoms (Fig. 1.2); the intermediate level $|4\rangle$ is nearly resonant with the pump waves. Kim and Coleman /51/ and Petuchowski et al. /52/, solved the density matrix equations in steady-state, and found three contributions to the Stokes polarization: first, a laser term proportional to the population inversion between levels $|2\rangle$ and $|3\rangle$; second, a Raman term proportional to the population difference between states $|4\rangle$ and $|3\rangle$ and to the intensity of the ν_1' -field; and third, the SHRS term proportional to the population difference between levels $|3\rangle$ and $|1\rangle$ and to the intensities of the ν_1 - and ν_1' -fields. Reasonable agreement has been found between measurement and theory in IR-IR and IR-Microwave pumped systems /51/.

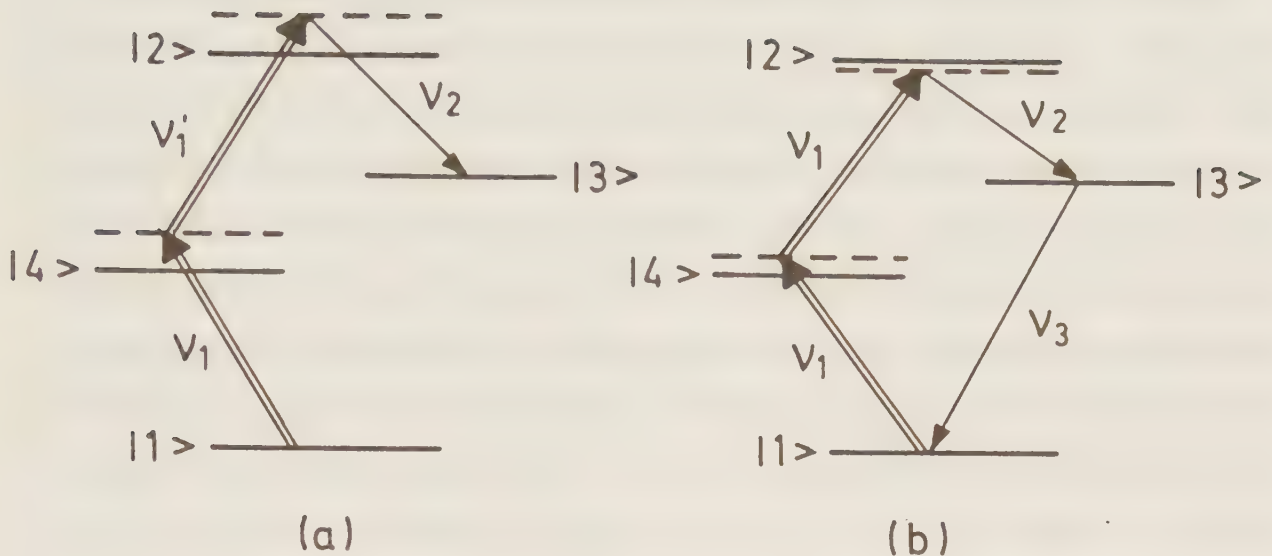


Fig. 1.2
Four level systems considered in theoretical studies
of SHRS: (a) Scheme used in [13, 51, 52], (b) Scheme
used in [14].

Hermann and Thompson [13] found analytical solutions for ultrashort-pulse propagation, when saturation effects are ignored in SHRS. They approached SHRS strictly as a three-photon resonant process, with the pumping assumed to be far away from resonance with both intermediate levels $|2\rangle$ and $|4\rangle$ (see Fig. 1.2a); therefore, their calculations relied on the assumption that the only relevant amplitudes are those of state $|1\rangle$ and $|3\rangle$. Most of the experimental work [10-12, 17, 51] has however been carried out taking advantage of the two-photon resonant enhancement, where the population of level $|2\rangle$ can be at least as important as that of level $|3\rangle$.

Another important effect neglected in references [13, 51, 52], is the idler field. In most experiments where the forward propagating SHRS has been observed, there is always present ultraviolet emission at the approximate frequency $2\nu_1 - \nu_2$. The build-up of the idler field introduces new nonlinearities into the development of the Stokes pulse.

Louisell and Meystre /14/ have considered the effect of both the two-photon resonance and the idler field. Motivated by an experiment of Reif and Walther /11/, they worked out a semiclassical theory of SHRS in an amplifier consisting of four-level atoms (see Fig. 1.2b) interacting with the pump, Stokes and idler fields. A numerical solution of the matter-field equations evidenced a significant depletion of the ground level, which leads to lasing and saturation of the Stokes field, and showed also that the only negligible population in the system is that of level $|3\rangle$. These findings point to the inadequacy of theories which neglect the population in level $|2\rangle$ to explain the available experimental data.

In experiments performed in the visible regime /9-12,17/, there is no preferred, nearly resonant intermediate level $|4\rangle$, so that all possible intermediate levels should be taken into account. At first sight this seems to increase the complexity of the system, however, we show that when all intermediate levels are far from resonance, the transition $|1\rangle \leftrightarrow |2\rangle$ behaves very much like a single photon transition, so that the number of active levels can be reduced to three (see fig 1.1b), the intermediate levels being adiabatically eliminated. Our three-level system differs however from the ordinary one in so far as all transitions are now allowed (one of them a 'two-photon transition').

The idler field, which arises from four-photon parametric conversion, couples levels $|1\rangle$ and $|3\rangle$, and is closely linked to the Stokes field. Since parametric conversion requires a certain degree of phase matching, it contributes only to the forward scattering, while SHRS contributes also to backward scattering. We will concentrate here on forward scattering, since it has been the most used experimental and theoretical configuration /9,11-17/. The idler field generation has been studied in some detail /12,15-17/ and measurements of the generated intensity as a

function of pump intensity as well as of atomic density are available. The corresponding nonlinear polarizations are usually given in terms of third-order susceptibilities as those applied in four-wave mixing. There is however a problem as regards the expected tuning behaviour of the idler field. Some authors have measured a strong line at fixed frequency /9,16/, while others claim that the strong idler component is tunable /12/. Both lines are actually to be expected, but their intensity ratio is not obvious. We address this problem by studying the gain/absorption spectrum of a weak probe and analyzing the transient polarizations after sudden changes in the pump field.

1.3 Previous results. - Second Harmonic Generation in gases.

Less understood is second harmonic generation (SHG) in gases. As already mentioned, optical SHG by electric dipole transitions (E1) is restricted to systems lacking a center of inversion. However, several methods have been suggested and applied during the last decade to destroy the symmetry of an atomic vapor and turn SHG into an allowed process. This was achieved by applying an external dc-electric field, so that one zero-frequency photon combines with two pump photons to give one photon of twice the pump frequency. This process, called dc-electric field induced SHG /18/, is described by a third order susceptibility and has been observed even with low power laser beams /19/.

If the higher order electric quadrupole (E2) and magnetic dipole (M1) interactions are considered, SHG can be obtained as a three-photon process, described by a dipole forbidden second-order susceptibility which can be made to radiate in the forward direction either by pumping with two noncollinear laser beams /20/, or by applying a dc-magnetic field to "rotate" the driven E2 or M1 moments /21/.

These processes are induced by external manipulation and can be satisfactorily explained. Somewhat surprising was however the observation of SHG when a single laser beam was focused into a metal vapor in the absence of any external field to break the symmetry /22-27,30/. This occurred even in the case of S-S transitions, which are non-relativistically forbidden to all multipole orders /25-27/.

The most popular theoretical model for this effect was suggested by Mossberg et al. /22/, and relies on the existence of spontaneously induced radial electrostatic fields due to photoionization. These fields arise from spatial charge separation following multiphoton ionization, and play the same role as an externally applied field in dc-electric field induced SHG. Bethune /29/ has analyzed in detail this process and other possible mechanisms which can occur with only neutral atoms present, and involve dipole-forbidden second order susceptibilities in focused beams. The radiating polarizations in this case are proportional to the gradient of the field. They are however negligible compared to those originating in photoionization, and even a small amount of ionization can already overwhelm their effect. In the presence of high power pulses, the probability for multiphoton ionization is large, and hence the spontaneously induced radial electric field model has become the favoured explanation of SHG in vapors.

None of the proposed models gives a completely satisfactory explanation of the experimental findings (see e.g. /24/). A series of measurements performed in highly excited states (Rydberg states) of Potassium /27/ motivated our interest in SHG. The formalism of this thesis is particularly suited to an analysis of several parametric dependences in SHG and to discuss the reliability of the different theoretical models.

1.4 Scope of the thesis.

The purpose of this thesis is the study of the nonlinear effects described in the preceding sections within the framework of the semiclassical approximation. That is, we describe the fields classically, and the atomic medium quantum mechanically in the density matrix formalism. In Chapter II, we describe a method allowing to derive the equations of motion for the relevant density matrix elements. It constitutes a perturbative adiabatic approximation for the amplitudes of the nonresonant intermediate states in terms of the amplitudes of the active resonant states. The nonresonant intermediate states are thus eliminated, leaving equations for the resonant amplitudes alone, and the description of the atomic system is reduced to that of a three-level system with all transitions allowed; the transition $|1\rangle \leftrightarrow |2\rangle$ is a two-photon transition, whereas the other ones are dipole transitions. A summary is given of the widely used Slowly Varying Envelope Approximation (SVEA), and the related field equations are derived. Backward propagating fields are neglected. The limitations on the range of validity of the model are given, and a comparison is made with two well-known systems, which are special cases of the one considered here, namely: the two-photon pumped two-level system, and the standard three-level system.

In the third Chapter, we discuss the steady-state properties of the system, its response to a weak signal/idler probe, and the propagation of Stokes and idler waves in both the linear and the saturation regimes.

Chapter four is devoted to an analysis of the signal transients after switching on or off the strong laser pump. In the first case we have optical nutation; in the second we study the free induction decay of the

system after steady-state preparation. Our calculation is close to that of Ducloy et al. /31/. In the sense that we study the response of the system to a weak signal probe, after sudden changes in the strong pump field. This procedure has the advantage of giving information in both the time and frequency domains simultaneously, and is often called time-frequency spectroscopy.

The numerical solution of the coupled Maxwell-density matrix equations for pulse propagation is given in Chapter five. Temporal and spatial build-up of the fields is shown for gaussian pump pulses in some particular cases. A comparison to recent measurements of the temporal location of Stokes and idler pulses with respect to the maximum of the pump pulse at different vapor pressures and pump intensities /12/ is made.

The sixth Chapter is devoted to a critical discussion of SHG in vapors. We show the importance of Stark shifts and Stark broadening, as well as that of phase matching. Considering that under typical experimental conditions collisions are also expected to play an important role, we have also included them in our analysis. We show that if an average dipole moment, independent of the atomic density and pump intensity, can be assigned to the forbidden transitions (S-S or S-D), all experimental curves can be reproduced. The average dipole moment can arise from mixing of the S,D states with P states; we discuss the predictions of the available theoretical models, consider the effect of I-mixing collisions, and estimate the corresponding conversion efficiencies for Potassium.

Finally in Chapter seven, we give a general discussion of our results, and suggest some topics as possibilities for future theoretical and experimental research.

CHAPTER II

THE MATTER-FIELD EQUATIONS

2.1 Introduction

Real atomic spectra include a denumerable set of discrete levels. Nevertheless, when interacting with a monochromatic electromagnetic field of frequency almost resonant with two of these levels, a drastic but surprisingly good approximation can be made: to treat the atomic system as consisting of precisely these two levels. The interaction of two-level atoms with electromagnetic fields has been investigated extensively in the past (for a review see e.g. the book by Allen and Eberly /32/). The vector model of Feynman, Vernon, and Hellwarth /33/, usually called the Bloch-vector model because of its similarity with the formalism used in NMR, has been a powerful tool for describing the coherent phenomena involved in the interaction between field and two-level system.

A similar approximation, that is, the consideration of $N+1$ levels only, can be made for N -photon processes where each field is almost resonant with an atomic transition. The construction of an adequate vector model becomes however more complicated, and has been carried out explicitly only for a three-level system interacting with two electromagnetic fields. This generalized vector model, due to Brewer and Hahn /36/, facilitates the study of two-photon coherent phenomena, and is an exact solution of the three-level problem in the absence of relaxations and when the input frequencies are resonant with the two-photon transition frequency. The interaction of three-level systems with two resonant fields has been widely discussed in the literature:

two-photon absorption, and Raman-type stimulated emission including line shape studies have been the object of detailed analysis by semiclassical methods /37-42/.

When an almost two-photon resonant electromagnetic field interacts with an atomic system, so that no atomic transition is resonant with the pump frequency, a direct two-photon absorption takes place and population is transferred from the ground state to the excited state. Intermediate states do not become significantly populated, and only contribute to the transition probability. As early as 1931, M. Goeppert-Mayer /43/ derived the probability for this process by means of second-order perturbation theory, but the observation of two-photon resonant absorption and its numerous applications had to wait for decades, until light sources intense enough became available.

In 1971 an approximate vector model for two-photon resonant transitions was introduced by Takatsuji /44/. It is an approximation which takes advantage of the fact, and therefore requires that, the frequency difference between the input light and the intermediate transitions be large. Grischkowsky et al. /45/ analyzed the similitudes of this model with the familiar one-photon model, and applied it to the study of two-photon resonantly enhanced parametric processes, such as third-harmonic generation, and of two-photon analogs of one-photon processes.

This two-photon vector model gives the same results as that of Brewer and Hahn for three-level systems, provided that only one intermediate level located very far from the one-photon resonance is considered /46/. It has however the advantages of including the effect of all possible intermediate levels, and it is not restricted to the case of exact two-photon resonance.

The standard method of finding equations of motion for slowly varying quantities is to transform into an interaction picture and then to apply the rotating wave approximation (see e.g. /47/). This method was however not used in ref./45/, nor will it be used here, because it leads to density matrix elements which are superpositions of terms oscillating rapidly at different frequencies. Grischkowsky et al. /45/ applied a different unitary transformation: the two active states are transformed into two new states, which include a linear superposition of the nonresonant intermediate states, and between which the electric dipole moment has non vanishing matrix elements. If the field (only one in their case, namely the pump field) is turned on and off adiabatically with respect to the intermediate levels, a two-photon vector model can be constructed: the system behaves then like a modified two-level system driven by an effective two-photon Rabi frequency and with a two-photon Stark shift proportional to the pump intensity.

When more than one field is associated with one atomic transition, as in our case, finding the appropriate unitary transformation becomes more complicated. Our procedure provides a way to overcome the problem. We expand the density matrix elements in the time-dependent Schroedinger equation in a Fourier series including all field frequencies and their combinations, and obtain differential equations for both resonant and nonresonant amplitudes. We solve then the equations for the nonresonant amplitudes in terms of the resonant ones by means of an adiabatic approximation, and finally find equations for the relevant slowly varying amplitudes alone. Our main approximation is closely related to that of Grischkowsky et al. /45/, and our model retains the characteristics of theirs as far as the two-photon transition is concerned. The detailed description of this multiple time perturbation technique, as well as the complete derivation of the equations for the

resonant density matrix elements, is given in Appendix 1. In the next paragraph we only mention the approximations involved and give the results of this procedure.

We then write the Maxwell equations in the Slowly Varying Envelope Approximation (SVEA), and conclude this Chapter by discussing analogies and differences between our system and the standard three-level system, as well as the range of validity of the approximations used throughout.

2.2 Density matrix equations

We consider the system of Fig. 2.1, with the three active atomic states labeled $|1\rangle$, $|2\rangle$, and $|3\rangle$, and the intermediate states ($|i\rangle$), $i=4, \dots$. States $|1\rangle$ and $|2\rangle$ have opposite parity to $|3\rangle$ and ($|i\rangle$). The atomic transition frequencies between levels $|j\rangle$, $|k\rangle$, are ω_{jk} . We assume the system to be homogeneously broadened (*) and to interact with three coherent fields E_1 , E_2 , E_3 , of frequencies ν_1 , ν_2 , and ν_3 respectively. The pump field frequency ν_1 is far from any one-photon resonance but is nearly two-photon resonant with frequency detuning $\Delta_1 = \omega_{21} - 2\nu_1$. The frequency detunings $\Delta_2 = \omega_{23} - \nu_2$, and $\Delta_3 = \omega_{31} - \nu_3$ are small enough so that we can associate the E_2 , E_3 fields with the atomic transitions $|2\rangle \leftrightarrow |3\rangle$, and $|1\rangle \leftrightarrow |3\rangle$ respectively. Furthermore we require conservation of energy, so that $2\nu_1 = \nu_2 + \nu_3$, and $\Delta_1 = \Delta_2 + \Delta_3$.

The electric field is assumed to be linearly polarized along $\underline{u}$ and given by:

(*) Frequency shifts due to the atomic motion can be readily included.

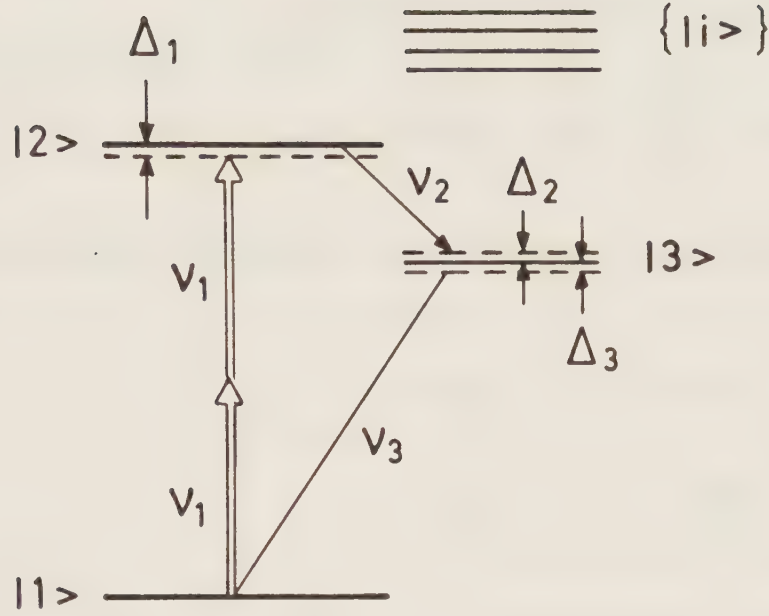


Fig. 2.1

Level scheme and field frequencies. The three active states are labeled $|1\rangle$, $|2\rangle$, $|3\rangle$. $\{li\rangle\}$ is the set of intermediate states. Δ_1 , Δ_2 , and Δ_3 are the two-photon, Stokes and idler detunings respectively.

$$E(z, t) = \sum_{\alpha=1}^3 E_{\alpha}(z, t) = 1/2 \sum_{\alpha=1}^3 \{ \tilde{E}_{\alpha}(z, t) \exp(ik_{\alpha}z - i\nu_{\alpha}t) + c.c. \} \quad (2.1)$$

where $k_{\alpha} = k_{\alpha} z$ is the wavevector in the medium. The complex field envelopes $\tilde{E}_{\alpha}(z, t)$ will be regarded as slowly varying, in amplitude as well as in phase, within a period and a wavelength of the carrier wave (SVEA), so that rapid time and space dependences are only contained in exponentials in Eq. (2.1). A discussion of the validity and the consequences of the SVEA will be given in the next Section.

The Hamiltonian of the system in the semiclassical approximation is

$$H = H_{\alpha} + U \quad (2.2)$$

where H_a is the atomic Hamiltonian in absence of fields, and $U = -\mathbf{d} \cdot \mathbf{E}(z, t)$ is the atom-field interaction in the dipole approximation. $\mathbf{d}$ is the atomic dipole operator $-e\mathbf{r}$. The interaction matrix elements $U_{jk} = \langle j|U|k \rangle$ are given by

$$U_{jk} = -d_{jk}/2 \sum_{\alpha=1}^3 (\tilde{\mathcal{E}}_{\alpha}(z, t) \exp(ik_{\alpha}z - i\nu_{\alpha}t) + \text{c.c.}) \quad (2.3)$$

where $d_{jk} = \langle j|\mathbf{d} \cdot \mathbf{u}|k \rangle$ is assumed to be real without loss of generality.

The density matrix evolves according to the equation

$$\frac{\partial \rho}{\partial t} = -i/\hbar [H, \rho] + \dot{\rho}_R \quad (2.4)$$

where relaxation processes such as spontaneous emission, collisional phase relaxation, or collisional population transfer, have been included phenomenologically in the second term. The corresponding matrix elements are usually given by

$$(\dot{\rho}_R)_{lj} = \langle l|\dot{\rho}_R|j \rangle = -\gamma_{lj} \rho_{lj} \quad \text{for } l \neq j \quad (2.5)$$

$$= -\Gamma_l \rho_{ll} + \sum_k \Gamma_{kl} \rho_{kk} \quad \text{for } l=j.$$

The off-diagonal relaxation rates γ_{lj} , or phase relaxation rates, are due mostly to elastic collisions. Those rates are usually larger than the population relaxation rates Γ_l and Γ_{kl} . We include both diagonal and off diagonal relaxation processes in one term by writing

$$(\dot{\rho}_R)_{lj} = \sum_{km} R_{ljkm} \rho_{km}, \quad \text{where } R_{ljkm} = -\gamma_{lj} \delta_{lk} \delta_{jm} (1 - \delta_{lj}) - \Gamma_l \delta_{lj} \delta_{lk} \delta_{lm} + \Gamma_{kl} \delta_{lj} \delta_{km} (1 - \delta_{lk}).$$

Substituting Eq. (2.2) into Eq. (2.4) yields then the equations of motion for the density matrix elements,

$$\dot{\rho}_{lj} = -i\omega_{lj}\rho_{lj} + (\dot{\rho}_R)_{lj} - i\hbar [U, \rho]_{lj} . \quad (2.6)$$

where $\omega_{ll} = 0$.

The matrix elements of ρ , as well as those of U , contain rapidly varying terms. We remove them by expanding ρ_{lj} in a Fourier series,

$$\rho_{lj}(t) = \sum'_{NLM} A_{lj}^{NLM} \exp(i\nu_{NLM}t) . \quad (2.7)$$

where A_{lj}^{NLM} are slowly varying amplitudes of rapidly oscillating terms with frequencies $\nu_{NLM} = N\nu_1 + L\nu_2 + M\nu_3$, where N, L, M are integers. Because of energy conservation, $2\nu_1 - \nu_2 - \nu_3 = 0$, we have

$$\nu_{NLM} = \nu_{N\pm 2, L\mp 1, M\mp 1} . \quad (2.8)$$

and therefore, care should be taken not to include the same frequency twice in the sum. This is indicated in Eq. (2.7) by the primed sum.

Since the density matrix elements satisfy $\rho_{lj} = \rho_{jl}^*$, the amplitudes A_{lj}^{NLM} must also satisfy

$$(A_{lj}^{NLM})^* = A_{jl}^{-N-L-M} . \quad (2.9)$$

Inserting Eq. (2.7) into Eq. (2.6) yields equations for the slowly varying amplitudes A_{lj}^{NLM} (see Eq. (A1.2)). For each ρ_{lj} , there is one resonant amplitude A_{lj}^{NLM} such that

$$\omega_{lj} + \nu_{NLM} \approx 0 . \quad (2.10)$$

and a large set of nonresonant ones.

We are interested in finding equations of motion for the resonant amplitudes. This can be achieved by solving equations (A1.2) for the nonresonant amplitudes under the following approximations:

- (1) The nonresonant amplitudes are usually smaller than the resonant ones, and are coupled only through the idler and signal fields. We neglect any coupling between them.
- (2) We are interested in retaining only first order terms in $\tilde{\mathcal{E}}_2$ and $\tilde{\mathcal{E}}_3$, on the basis that the pump field is much stronger than idler and signal. Terms coupling nonresonant amplitudes with resonant ones through the signal/idler fields give rise to second order terms in those fields; therefore we will neglect them. It is shown in Appendix 1 that crossed terms $\tilde{\mathcal{E}}_1\tilde{\mathcal{E}}_2$ or $\tilde{\mathcal{E}}_1\tilde{\mathcal{E}}_3$ do not couple two resonant amplitudes, and will not appear in the final equations.
- (3) We assume that the temporal change of the amplitude $\tilde{\mathcal{E}}_1(z,t)$ is slow on a time scale given by the inverse of any intermediate detuning, i.e., that the pump field is adiabatically applied with respect to any intermediate state. This condition can always be satisfied by choosing the pump frequency far enough from any intermediate resonance. For one-photon pump detunings in the visible this condition will be fulfilled for pulses as short as picosecond, and in the near infrared down to nanosecond. This is the main assumption leading to the elimination of the intermediate states in two-photon pumped systems.

Under these conditions, and removing also the rapidly varying terms in space, one obtains equations for the resonant slowly varying amplitudes $\tilde{\mathcal{P}}_j$ (see Appendix 1):

$$\begin{aligned}
\dot{\tilde{\rho}}_{21} &= -(\gamma_{21} + i\Delta_1 + i\omega_{s1}I_1)\tilde{\rho}_{21} + (i\tilde{\rho}_{23}\tilde{V}_{31} - i\tilde{V}_{23}\tilde{\rho}_{31})\exp(-i\Delta kz) + i\tilde{V}_{21}(\tilde{\rho}_{22} - \tilde{\rho}_{11}) \\
\dot{\tilde{\rho}}_{23} &= -(\gamma_{23} + i\Delta_2 + i\omega_{s2}I_1)\tilde{\rho}_{23} + (i\tilde{\rho}_{21}\tilde{V}_{13} - i\tilde{V}_{21}\tilde{\rho}_{13})\exp(i\Delta kz) + i\tilde{V}_{23}(\tilde{\rho}_{22} - \tilde{\rho}_{33}) \\
\dot{\tilde{\rho}}_{13} &= -(\gamma_{13} - i\Delta_3 - i\omega_{s3}I_1)\tilde{\rho}_{13} + (i\tilde{\rho}_{12}\tilde{V}_{23} - i\tilde{V}_{12}\tilde{\rho}_{23})\exp(-i\Delta kz) - i\tilde{V}_{13}(\tilde{\rho}_{33} - \tilde{\rho}_{11}) \\
\dot{\tilde{\rho}}_{11} &= \gamma_3\tilde{\rho}_{33} + (i\tilde{\rho}_{12}\tilde{V}_{21} + i\tilde{V}_{31}\tilde{\rho}_{13} + \text{c.c.}) \\
\dot{\tilde{\rho}}_{22} &= -\gamma_2\tilde{\rho}_{22} - (i\tilde{V}_{21}\tilde{\rho}_{12} + i\tilde{\rho}_{32}\tilde{V}_{23} + \text{c.c.}) \\
\dot{\tilde{\rho}}_{33} &= -\gamma_3\tilde{\rho}_{33} + \gamma_2\tilde{\rho}_{22} + (i\tilde{\rho}_{32}\tilde{V}_{23} + i\tilde{V}_{13}\tilde{\rho}_{31} + \text{c.c.})
\end{aligned} \tag{2.11}$$

where the reduced interaction matrix elements $\tilde{V}_{ij}$ are given by

$$\begin{aligned}
\tilde{V}_{23} &= -d_{23}\tilde{\mathcal{E}}_2/2\hbar \\
\tilde{V}_{31} &= -d_{31}\tilde{\mathcal{E}}_3/2\hbar \\
\tilde{V}_{21} &= k_{21}\tilde{\mathcal{E}}_1^2/2\hbar.
\end{aligned} \tag{2.12}$$

and k_{21} is the effective two-photon coupling constant

$$k_{21} = 1/2\hbar \sum_j d_{2j}d_{j1}/(\omega_{2j} - \nu_1). \tag{2.13}$$

The two-photon Stark shifts $\omega_{si}I_i$, $i=1,2,3$, have been expressed in terms of the dimensionless intensity $I_i = 2|V_{21}| \sqrt{T_i/\gamma_{21}}$ with $T_i = 1/2 (2/\gamma_2 + 1/\gamma_3)$ and

$$\omega_{s1} = -1/2\hbar \sum_j \frac{|d_{1j}|^2 \omega_{1j}}{\omega_{1j}^2 - \nu_1^2} + \frac{|d_{2j}|^2 \omega_{j2}}{\omega_{j2}^2 - \nu_1^2} (1/k_{21} \sqrt{\gamma_{21}/T_1})$$

$$\omega_{s2} = -1/2\hbar \sum_j \frac{|d_{3j}|^2 \omega_{3j}}{\omega_{3j}^2 - \nu_1^2} + \frac{|d_{j2}|^2 \omega_{j2}}{\omega_{j2}^2 - \nu_1^2} (1/k_{21} \sqrt{\gamma_{21}/T_1})$$

$$\omega_{s3} = \omega_{s1} - \omega_{s2}. \quad (2.14)$$

The exponential factor $\exp(i\Delta kz)$ in Eq. (2.11), with $\Delta k = 2k_1 - k_2 - k_3$, describes the effect of phase mismatch for four-photon parametric processes in the plane wave approximation.

To obtain a closed system of equations, the following assumptions have been made about the relaxation times: $|1\rangle$ is the ground state, $|2\rangle$ decays mainly into $|3\rangle$, so that $\Gamma_2 = \Gamma_{23} \equiv \gamma_2$, and $|3\rangle$ into $|1\rangle$ so that $\Gamma_3 = \Gamma_{31} \equiv \gamma_3$. All other population relaxation rates have been neglected. In such a closed system the trace of the density matrix is conserved.

2.3 The Slowly Varying Envelope Approximation (SVEA)

From Maxwell equations for homogeneous dielectric media, one obtains the wave equation:

$$\nabla^2 \underline{E} - \frac{1}{c^2} \frac{\partial^2 \underline{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \underline{P}}{\partial t^2}. \quad (2.15)$$

Following the same procedure as for the fields (Eq. (2.1)), we express the polarization $\underline{P}(z, t)$ as a superposition of quasi-monochromatic waves:

$$\underline{P}(z, t) = \sum_{\alpha=1}^3 \underline{P}_{\alpha}(z, t) = 1/2 \sum_{\alpha=1}^3 \{ \tilde{\underline{P}}_{\alpha}(z, t) \exp(ik_{\alpha} z - i\nu_{\alpha} t) + \text{c.c.} \}. \quad (2.16)$$

We write the polarization as the sum of its linear and nonlinear contributions

$$P_{\alpha} = P_{\alpha}^L + P_{\alpha}^{NL} . \quad (2.17)$$

where the linear polarization P_{α}^L is given by

$$P_{\alpha}^L = \epsilon_0 N \chi(\nu_{\alpha}) E_{\alpha} . \quad (2.18)$$

with N the atomic density, and $\chi(\nu_{\alpha})$ the first-order susceptibility. $\chi(\nu_{\alpha})$ is usually a tensor but for isotropic media and linearly polarized waves $E = E \underline{u}$, Eq. (2.15) and Eq. (2.18) become scalar equations. By inserting the linear index of refraction n_{α} , defined by

$$n_{\alpha}^2 - 1 = N \operatorname{Re} \chi(\nu_{\alpha}) , \quad (2.19)$$

in Eq. (2.15), one obtains

$$\frac{\partial^2 E_{\alpha}}{\partial z^2} - \frac{1}{v_{\alpha}^2} (1 + i \frac{N \operatorname{Im}(\chi)}{n_{\alpha}^2}) \frac{\partial^2 E_{\alpha}}{\partial t^2} = \mu_0 \frac{\partial^2 P_{\alpha}^{NL}}{\partial t^2} . \quad (2.20)$$

where $v_{\alpha} = c/n_{\alpha}$ is the phase velocity in the medium. The linear polarization is thus incorporated in the left hand side of Eq. (2.20). We now introduce the SVEA, which yields first-order differential equations for the fields.

Maxwell equations in the SVEA are obtained from Eq. (2.20) by assuming that, as the wave propagates through the medium, its amplitude and phase vary slowly within a wavelength and a period of the carrier. The nonlinear polarization amplitudes are also required to be slowly

varying in time, i.e.,

$$\left| \frac{\partial^2 \tilde{E}_\alpha}{\partial z^2} \right| \ll \left| k_\alpha \frac{\partial \tilde{E}_\alpha}{\partial z} \right|, \quad \left| \frac{\partial^2 \tilde{E}_\alpha}{\partial t^2} \right| \ll \left| \nu_\alpha \frac{\partial \tilde{E}_\alpha}{\partial t} \right|,$$

$$\left| \frac{\partial^2 \tilde{\rho}_\alpha^{NL}}{\partial t^2} \right| \ll \left| \nu_\alpha \frac{\partial \tilde{\rho}_\alpha^{NL}}{\partial t} \right| \ll \left| \nu_\alpha^2 \tilde{\rho}_\alpha^{NL} \right|. \quad (2.21)$$

Under these conditions, the wave equation reduces to

$$\frac{\partial \tilde{E}_\alpha}{\partial z} + \frac{1}{\nu_\alpha} \left(1 + \frac{i N \text{Im} \chi}{n_\alpha^2} \right) \frac{\partial \tilde{E}_\alpha}{\partial t} + a_\alpha \tilde{E}_\alpha = \frac{i k_\alpha}{2 \epsilon_\alpha} \tilde{\rho}_\alpha^{NL}. \quad (2.22)$$

where the absorption coefficient a_α is given by

$$a_\alpha = \frac{N \text{Im}(\chi) k_\alpha}{2 n_\alpha^2} \quad (2.22a)$$

and $\epsilon_\alpha = n_\alpha^2 \epsilon_0$. The macroscopic polarization P is obtained from the atomic dipole moments as

$$P = N \text{Tr}(\rho d) = N \sum_j \rho_{lj} d_{jl}. \quad (2.23)$$

From equations (2.23), (2.16) and (2.7), we obtain expressions for the slowly varying polarizations in terms of the density matrix elements; this procedure yields resonant and nonresonant contributions to $\tilde{\rho}_\alpha$, $\alpha=1,2,3$. The nonresonant polarizations can be calculated from Eq. (A1.2), by restricting the signal and idler fields through adiabatic

criteria similar to that of Eq. (A1.8) (*). They give rise to the linear polarization as well as to a small contribution subjected to phase matching, so that the total polarization reads

$$\tilde{P}_1 = N\epsilon_0\chi(\nu_1)\tilde{E}_1 - 4Nk_{21}\tilde{P}_{21}\tilde{E}_1^* \quad (2.24a)$$

$$\tilde{P}_2 = N\epsilon_0\chi(\nu_2)\tilde{E}_2 + 2N\kappa\tilde{P}_{21}\tilde{E}_3^* \exp(i\Delta kz) + 2Nd_{23}\tilde{P}_{23} \quad (2.24b)$$

$$\tilde{P}_3 = N\epsilon_0\chi(\nu_3)\tilde{E}_3 + 2N\kappa\tilde{P}_{21}\tilde{E}_2^* \exp(i\Delta kz) + 2Nd_{13}\tilde{P}_{21} . \quad (2.24c)$$

The first two terms in Eqs. (2.24a,b,c) come from nonresonant polarizations, while the last term in Eqs. (2.24b,c) comes from the resonant contribution. The susceptibility $\chi(\nu_\alpha)$ and the factor κ are given by

$$\chi(\nu_\alpha) = 1/\epsilon_0\hbar \sum_{lj} \frac{d_{lj} d_{jl} (P_{jj} - P_{ll}) f_{lj}^{(\omega)}}{\omega_{lj} - \nu_\alpha - i\gamma_{lj}} , \quad \alpha = 1, 2, 3 \quad (2.25)$$

and

$$\kappa = 1/2\hbar \sum_l d_{1l} d_{l2} \frac{2(\omega_{l1} - \nu_1) - (\omega_{l1} - \nu_2) \delta_{l3}}{(\omega_{l1} - \nu_2)(\omega_{l1} - \nu_3)} . \quad (**) \quad (2.26)$$

(*) The field envelopes must be slowly varying in a time scale given by the inverse of the detuning to any nonresonant state: (see Appendix 1 for notation)

$$\begin{aligned} |\tilde{E}_2| &>> \left| \frac{1}{\Delta_{lj}^{0-10}} \frac{\partial \tilde{E}_2}{\partial t} \right| , \quad lj \neq 23 . \\ |\tilde{E}_3| &>> \left| \frac{1}{\Delta_{lj}^{0-1}} \frac{\partial \tilde{E}_3}{\partial t} \right| , \quad lj \neq 31 . \end{aligned} \quad (A1.18)$$

(**) For k_{21} (Eq. 2.13) and κ (Eq. 2.26) in Eqs. (2.24) we assumed $\gamma_{lj} \sim \gamma_{2l} \ll \omega_{l1} - \nu_\alpha$, $\alpha = 1, 2, 3$.

The factors $f_{lj}^{(2)} = 1 - \delta_{lj,23}$, $f_{lj}^{(3)} = 1 - \delta_{lj,31}$, are introduced to exclude explicitly the lj terms corresponding to the resonant polarization from the sum; there is no resonant polarization oscillating at the pump frequency, therefore $f_{lj}^{(1)} = 1$. The first term in Eqs. (2.24a,b,c) has the same form as the linear polarization P_α^L defined in Eq. (2.18). For the pump field it corresponds to the linear polarization, as defined within perturbation theory. For the Stokes and idler it contains only the contribution of the nonresonant background to the linear polarization, in so far as the resonant contribution is explicitly excluded through the $f_{lj}^{(\alpha)}$ factors (*).

The term proportional to $\exp(i4kz)$ in Eqs. (2.24) can be neglected compared with the resonant contribution, because of the smallness of χ (Eq. (2.26)). Also, if γ_{lj} in Eq. (2.25) is much smaller than $\omega_{lj} - \nu_\alpha$, $\text{Im}(\chi) \ll \text{Re}(\chi)$ and the background absorption (determined by the coefficient a_α (Eq. (2.22a))) is negligible and the field equations (2.22) may be expressed as

$$\frac{\partial \tilde{E}_1}{\partial z} + \frac{1}{v_1} \frac{\partial \tilde{E}_1}{\partial t} = ik_1 N / \epsilon_1 (-2k_{21} \tilde{P}_{21} \tilde{E}_1^*) \quad (2.27a)$$

$$\frac{\partial \tilde{E}_2}{\partial z} + \frac{1}{v_2} \frac{\partial \tilde{E}_2}{\partial t} = ik_2 N / \epsilon_2 d_{32} \tilde{P}_{23} \quad (2.27b)$$

$$\frac{\partial \tilde{E}_3}{\partial z} + \frac{1}{v_3} \frac{\partial \tilde{E}_3}{\partial t} = ik_3 N / \epsilon_3 d_{13} \tilde{P}_{31} \quad (2.27c)$$

For convenience, we omit in the following chapters the tilde over $\tilde{P}$, $\tilde{V}$

(*) In the usual expression for the index of refraction, it is assumed that most of the atoms are in the ground state. Near a resonance the same first order expression (2.25) for $\chi^{(1)}(\nu_\alpha)$ is used (omitting the factors $f_{lj}^{(\alpha)}$), therefore γ_{lj} becomes of crucial importance.

and $\mathcal{E}$.

2.4 Discussion of the method

Equations (2.11) correspond to a three-level system having all transitions allowed, one of them a two-photon transition. By switching off the pump field, we recover the standard three-level system equations in the RWA (see e.g. /36/), and by switching off the signal and idler fields, we recover the two-photon two-level system of Grischkowsky et al. /45/. The approximate two-photon vector model allows for resonant and nonresonant two-photon transitions, and is valid for any number of intermediate states, whenever the adiabatic criterion of Eq. (A1.8) is satisfied for each one. We emphasize here that this adiabatic criterion refers to intermediate transitions, that is, the pump field should be adiabatically applied only with respect to the intermediate states, but not necessarily with respect to the two-photon resonant level.

As already pointed out by Grischkowsky et al. /45/, the two-photon vector model is useful in the study of two-photon analogs of well-known one-photon processes, like photon echoes, optical nutation, adiabatic following, adiabatic rapid passage, etc.; however the presence of Stark shifts in Eq. (2.11), which are pump intensity dependent detunings, gives rise to more complicated effects. The characteristics of the two-photon two-level model are retained in our model, as well as those of a standard three-level system interacting with the signal and idler fields, but they can interfere and give rise to new effects. A detailed analysis of the steady-state response of the system, as well as of its transient behaviour, is presented in the next Chapters.

We conclude this Chapter by summarizing the approximations involved in the derivation of our working equations:

- (1) As already mentioned, the coupling between nonresonant amplitudes is neglected. This coupling arises from the signal and idler fields. By inspection of Eq. (A1.7) we see that if it is included, to apply the same method for solving the integral equation (A1.6) would require the signal and idler fields to satisfy also an adiabatic criterion. The coupling terms are of the order of $B_{km}^{NLN} \tilde{\epsilon}_{23}$, and can be neglected if $|V_{23}|, |V_{13}| \ll \Delta_{km}^{NLN}$.
- (2) Second-order terms in signal/idler fields are neglected: they are assumed to be much weaker than the pump field, so that they do not excite off-resonant transitions.
- (3) The adiabatic condition, Eq. (A1.8), for the nonresonant amplitudes is applied. For the amplitudes B_{ij}^{100} , this condition reduces to that of Grischkowsky et al. /45/, but it is actually more general and restrictive here because it applies to all nonresonant amplitudes B_{kj}^{NLN} . (*)

It is worthwhile making a last comment on the assumption of nondegenerate states. In real systems, each J-level is $(2J+1)$ -fold degenerate. For linearly polarized fields, only transitions with $\Delta M=0$ are allowed, and each M-subsystem can be treated separately and the results added to obtain the behaviour of the total system, as a

(*) In Eq. (A1.12) the amplitudes B_{km}^{NLN} with $k \geq 3$ are seen to contribute to the two-photon effective matrix element V_{21} , coupling ρ_{23}^{c-10} and ρ_{13}^{c01} . In an earlier publication /34/, only the B_{33}^{NLN} amplitude was considered and nonresonant polarizations between intermediate states were neglected. This led to the introduction of a partial two-photon matrix element V_{21R} including only the term $k=3$ in the sum defining V_{21} . B_{33}^{NLN} corresponds to a nonresonant population in the intermediate state, which is expected to be important, while B_{k3}^{NLN} , $k \geq 3$, are amplitudes for nonresonant polarizations between intermediate states, oscillating at frequencies $\pm \Delta$, $\pm (2\nu_1 \pm \Delta)$, where $\Delta = \nu_2 - \nu_1$. Their contribution is however of the same order of magnitude as that of B_{33}^{NLN} . The need to consider these terms was pointed out by H. G. von Garssen /35/, who developed a computer code to determine systematically all frequency combinations required to properly describe the evolution of the atomic density matrix. The equations (A1.14) were obtained as a first order approximation. The computer code gives also the possibility of extending the method to higher orders.

superposition of the noninteracting M-subsystems. When M-changing collisions can not be neglected, or when fields of different polarizations act on the system, Eq. (2.6) must be solved simultaneously for all M-subsystems, and the calculation becomes much more complicated. To avoid this complication, the present work is limited to linearly polarized fields.

CHAPTER III

STEADY STATE REGIME

3.1 Introduction

The steady state analysis of the interaction between a nonlinear medium and electromagnetic fields is interesting in many respects. First, and obviously, it gives the response of the system to steady-state input fields -this, however, is not very realistic for cases such as discussed here, since the necessary intensities can be reached in practice only with pulsed lasers. A second case of interest is that of very fast atomic relaxation times, in which case the medium follows adiabatically the field variations. Finally, steady-state solutions are very useful in providing the physical intuition required to understand numerical results.

In this chapter, we consider the steady-state behavior of the atomic system subjected to an arbitrarily strong pump field and a weak probe field almost at resonance with the $|2\rangle \leftrightarrow |3\rangle$ or $|1\rangle \leftrightarrow |3\rangle$ transition (see Fig. 2.1), as well as competition effects in the build-up of these fields. We solve the density matrix equations to first order in the probe fields, and obtain the so-called weak probe gain from Maxwell's equations. Analytical expressions for the propagating fields in the amplifier are obtained, and some particular cases of interest are analyzed in detail.

Because of the large gain associated with the $|2\rangle \leftrightarrow |3\rangle$ transition, saturation of the corresponding Stokes field is usually reached quickly. We handle this situation by solving the density matrix equations to

second order in this field. This yields an analytical expression for the saturation intensity, as well as some insight into the behavior of the system in the saturation regime.

We compare our results with the measurements performed by Heinrich in Cadmium vapor /12/ and conclude this chapter by a brief discussion of phase mismatch effects.

3.2 Steady-state solution in the linear regime

The steady-state regime is obtained from Eqs. (2.11) by setting all temporal derivatives to zero. We obtain a system of six algebraic equations:

$$\begin{aligned}
 \rho_{21} &= D_1 \{ (i \rho_{23} V_{31} - i V_{23} \rho_{31}) \exp(-i \Delta k z) + i V_{21} (\rho_{22} - \rho_{11}) \} \\
 \rho_{23} &= D_2 \{ (i \rho_{21} V_{13} - i V_{21} \rho_{13}) \exp(i \Delta k z) + i V_{23} (\rho_{22} - \rho_{33}) \} \\
 \rho_{13} &= D_3 \{ (i \rho_{12} V_{23} - i V_{12} \rho_{23}) \exp(-i \Delta k z) - i V_{13} (\rho_{33} - \rho_{11}) \} \\
 \rho_{22} &= (-1/\gamma_2) (i \rho_{12} V_{21} + i V_{23} \rho_{32} + \text{c.c.}) \\
 \rho_{33} &= (1/\gamma_3) \{ (\gamma_2 \rho_{22} + (i \rho_{32} V_{23} + i V_{13} \rho_{31} + \text{c.c.})) \} \\
 \rho_{11} + \rho_{22} + \rho_{33} &= 1 .
 \end{aligned} \tag{3.1}$$

where we have introduced the complex Lorentzians

$$\begin{aligned}
 D_1 &= (\gamma_{21} + i \Delta_1 + i \omega_{s1} I_1)^{-1} \\
 D_2 &= (\gamma_{23} + i \Delta_2 + i \omega_{s2} I_1)^{-1} \\
 D_3 &= (\gamma_{13} - i \Delta_3 - i \omega_{s3} I_1)^{-1} .
 \end{aligned} \tag{3.1a}$$

We solve this system of equations in the limit of weak probe fields, i.e. for weak coupling energies V_{23} and V_{13} . We proceed perturbatively by first obtaining the zeroth order solution, given by $V_{23} = V_{13} = 0$, and then solving for the coherences ρ_{23} and ρ_{13} to first order. As readily seen from the Maxwell's wave equation (2.27b,c), these give directly the gain coefficients for the signal and idler fields. We obtain

$$\rho_{22} = \frac{\mathcal{L}_1 I_1^2}{2\gamma_2 T_1 S_1} \quad (3.2a)$$

$$\rho_{33} = (\gamma_2/\gamma_3) \rho_{22} \quad (3.2b)$$

$$\rho_{11} = 1 - \rho_{22} - \rho_{33} \quad (3.2c)$$

$$\rho_{21} = -iD_1 \dot{V}_{21}/S_1 \quad (3.2d)$$

$$\rho_{23} = \frac{D_2}{S_1 S_{23}} \{ iV_{23} (\beta + D_3 D_1^* |V_{21}|^2) + V_{21} V_{13} (D_1 + D_3(\beta+1)) \exp(i\Delta kz) \} \quad (3.2e)$$

$$\rho_{13} = \frac{D_3}{S_1 S_{23}} \{ iV_{13} (1 + \beta - D_2 D_1 |V_{21}|^2) + V_{12} V_{23} (D_2 \beta - D_1^*) \exp(-i\Delta kz) \}. \quad (3.2f)$$

Here we have introduced the Stark-shifted Lorentzian

$$\mathcal{L}_1 = \frac{\gamma_{21}^2}{\gamma_{21}^2 + (\Delta_1 + \omega_{S1} I_1)^2}$$

as well as the saturation denominators

$$S_1 = 1 + \mathcal{L}_1 I_1^2$$

$$S_{23} = 1 + D_2 D_3 |V_{21}|^2.$$

The coefficient β is given by

$$\beta = \rho_1 I_1^2 (1/\chi_2 - 1/\chi_3)/2T_1 .$$

3.3 Field propagation

Substituting the expressions (3.2d,e,f) for the coherences ρ_{21} , ρ_{23} , and ρ_{13} into Maxwell's Eqs. (2.27) gives a system of three coupled differential equations which describe the propagation of the pump, Stokes, and idler fields $\mathcal{E}_1$, $\mathcal{E}_2$, and $\mathcal{E}_3$ through the medium. At this point it is convenient to introduce the Rabi frequencies:

$$\Omega_{21} = k_{21} \mathcal{E}_1^2 / 2\hbar$$

$$\Omega_{23} = d_{23} \mathcal{E}_2 / 2\hbar$$

$$\Omega_{13} = d_{13} \mathcal{E}_3^* / 2\hbar$$

Note that the pump field induces a two-photon excitation and must thus be described by an effective two-photon Rabi frequency Ω_{21} . Eqs. (2.27) become

$$\frac{d\Omega_{21}}{dz} = -2i\alpha_1 \rho_{21} - |\Omega_{21}| \quad (3.3a)$$

$$\frac{d\Omega_{23}}{dz} = i\alpha_2 \rho_{23} = A\Omega_{23} + B\Omega_{13} \exp(i\Delta kz) \quad (3.3b)$$

$$\frac{d\Omega_{13}}{dz} = -i\alpha_3 \rho_{13} = C\Omega_{13} + D\Omega_{23} \exp(-i\Delta kz) . \quad (3.3c)$$

where

$$\alpha_1 = \frac{2k_1 N}{\epsilon_1} k_{21} \quad (3.4a)$$

$$\alpha_2 = \frac{k_2 N}{\epsilon_2} \frac{d_{23}^2}{2\hbar} \quad (3.4b)$$

$$\alpha_3 = \frac{k_3 N}{\epsilon_3} \frac{d_{13}^2}{2\hbar} \quad (3.4c)$$

$$A = \alpha_2 \frac{D_2}{S_1 S_{23}} (\beta + D_2 D_1^* |V_{21}|^2) \quad (3.4d)$$

$$B = -i\alpha_2 \frac{D_2}{S_1 S_{23}} V_{21} (D_1 + D_3 (\beta + 1)) \quad (3.4e)$$

$$C = -\alpha_3 \frac{D_3}{S_1 S_{23}} (1 + \beta - D_2 D_1 |V_{21}|^2) \quad (3.4f)$$

$$D = i\alpha_3 \frac{D_3}{S_1 S_{23}} V_{12} (D_2 \beta - D_1^*) \quad (3.4g)$$

Equation (3.3a) describes pump depletion which can be ignored provided that $\alpha_1 L \ll 1$, where L is the amplifier length and

$$\alpha_1 \sim 2\hbar k_{21} (\alpha_2/d_{23}^2 + \alpha_3/d_{13}^2).$$

As this is usually the case, we also neglect pump depletion in this analysis (in particular, no pump depletion was observed in the experiments of Refs. /12/ and /17/). The phase matching factors $\exp(\pm i\Delta kz)$ in Eqs. (3.3b) and (3.3c) can be eliminated by introducing the new variable

$$\Omega_{13}' = \Omega_{13} \exp(i\Delta kz),$$

so that Maxwell's equations reduce to the coupled-mode equations

$$\frac{d\Omega_{23}}{dz} = A \Omega_{23} + B \Omega_{13}' \quad (3.5a)$$

$$\frac{d\Omega_{13}'}{dz} = C' \Omega_{13}' + D \Omega_{23}. \quad (3.5b)$$

where

$$C' = C + i\Delta k. \quad (3.5c)$$

Equations (3.5) can easily be solved by Laplace transform technique, giving

$$\Omega_{23} = \frac{[B \Omega_{13}^0 + (r_+ - C') \Omega_{23}^0] e^{r_+ z} - [\Omega_{13}^0 B + (r_- - C') \Omega_{23}^0] e^{r_- z}}{r_+ - r_-}, \quad (3.6a)$$

$$\Omega_{13}' = \frac{[D \Omega_{23}^0 + (r_+ - A) \Omega_{13}^0] e^{r_+ z} - [D \Omega_{23}^0 + (r_- - A) \Omega_{13}^0] e^{r_- z}}{r_+ - r_-}, \quad (3.6b)$$

where

$$r_{\pm} = \frac{A + C' \pm \sqrt{(A - C')^2 - 4DB}}{2} \quad (3.6c)$$

and Ω_{23}^0 and Ω_{13}^0 are the initial Rabi frequencies of the Stokes and idler fields at $z=0$.

We can gain some insight into Eqs. (3.6) by analyzing particular cases. If we assume that γ_2 is small compared with the transverse relaxation constants (long lived upper level), and consider pump fields

weak enough to insure that $V_{21} D_{2,2} < 1$ (*), then we can neglect both the product 4DB inside the root in Eq. (3.6c) and the $D_2 D_1^*$, $D_2 D_1$ terms in Eqs. (3.4d) and (3.4f). This gives $r_+ \sim A$, $r_- \sim C'$. Furthermore, if only a probe field $\mathcal{E}_2$ is applied, so that $\Omega_{23}^0 \neq 0$ but $\Omega_{13} = 0$, then

$$\Omega_{23} = \Omega_{23}^0 \exp(Az) \quad (3.7a)$$

$$\Omega_{13} = \Omega_{23}^0 \{D/(A-C')\} (\exp(Az) - \exp(C'z)) \quad (3.7b)$$

In this case the Stokes probe field is exponentially amplified with gain A . From Eqs. (3.4d), (3.5c) and (3.4f) one finds that the real part of C' has a sign opposite to that of A . Thus the second exponential in equation (3.7b) decays away rapidly and the growth of the idler field is essentially also given by the Stokes gain. We note that the main effect of phase mismatch is to reduce the factor $D/(A-C')$ and we will disregard it in this case.

Note that if only a probe field $\mathcal{E}_3$ is applied, $\Omega_{13}^0 \neq 0$ and $\Omega_{23} = 0$. In this case, we obtain instead of (3.7)

$$\Omega_{23} = \Omega_{13}^0 \{B/(A-C)\} (\exp(Az) - \exp(Cz)) \quad (3.8a)$$

$$\Omega_{13} = \Omega_{13}^0 \exp(Cz) \quad (3.8b)$$

This indicates that the weak probe field $\mathcal{E}_3$ is absorbed, while the Stokes field still increases exponentially with z . The different behavior of the idler and signal fields is caused by the population distribution. In the absence of a probe field we have most of the population in the ground state, some in the upper level $|2\rangle$, and much

(*) The field can still be a saturating field for the two-photon transition, i.e., $|V_{21}| > \sqrt{\hbar\gamma_{21}/T_1}$.

less in level $|3\rangle$. That is, we have population inversion between levels $|2\rangle$ and $|3\rangle$, leading to lasing and idler field generation. A probe coupled to the $|1\rangle \leftrightarrow |3\rangle$ transition experiences essentially absorption.

We now concentrate on the analysis of the case, when a Stokes probe is applied. From Eqs. (3.7), we obtain readily that the growth of the field intensities is given by

$$I_2 = I_2^0 \exp(\alpha_2' z) \quad (3.9a)$$

$$I_3 = I_2^0 \left| \{D/(A-C)\} \{\exp(Az) - \exp(Cz)\} \right|^2, \quad (3.9b)$$

where

$$\alpha_2' = \frac{\alpha_2 \left(\frac{1}{T_2} - \frac{1}{T_3} \right) \rho_1 I_1^2}{2T_1 S_1} \left(\frac{D_2}{1 + D_2 D_3 |V_{21}|^2} + \text{c.c.} \right). \quad (3.9c)$$

Fig. 3.1 shows the Stokes and idler intensities as functions of the amplifier coordinate z for parameters under unsaturated gain conditions. Parameters chosen correspond to the levels $5s^2 {}^1S_0$, $5s6s {}^1S_0$, $5s5p {}^1P_1$ of Cd [12]. For simplicity, Stark shifts and detunings are set equal to zero. Dashed lines correspond to the analytical solution given by Eq. (3.9) for weak Stokes field, and the full ones to the numerical solution of Eqs. (3.5) for arbitrarily intense Stokes field. We see that the analytical approach gives a good description of the growth of the fields for intensities I_2 smaller than the saturation value I_2^{sat} also shown in the figure. (I_2^{sat} is defined precisely in Section 3.5). The dip in the idler intensity is due to a change of sign in ρ_{13} , which leads to a zero in the Rabi frequency. It occurs at the transition between the small signal region, where both Stokes and idler fields are generated as

a single four-photon process, starting and ending in the ground state, and the region where the Stokes field becomes intense enough to inhibit this process, by establishing Rabi oscillations between level $|2\rangle$ and $|3\rangle$.

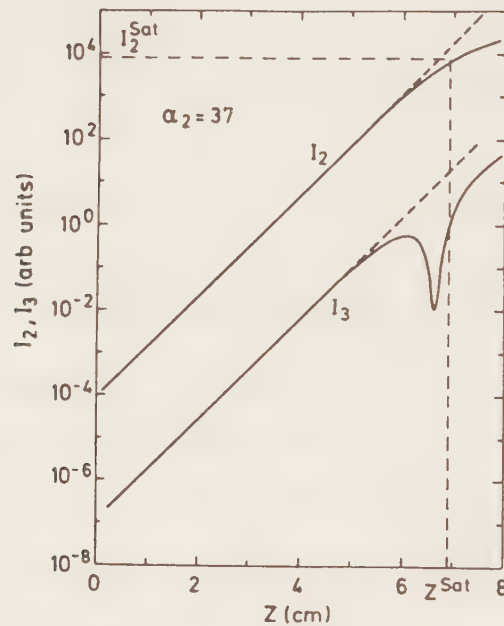


Fig. 3.1

Stokes (I_2) and idler (I_3) intensities as functions of the amplifier coordinate z . The Stokes saturation intensity I_2^{Sat} indicates the end of the small gain regime. Relaxation rates are $\gamma_2=0.1$, $\gamma_3=1$, $\gamma_{21}=\gamma_{23}=\gamma_{13}=10$; $\alpha_3=10\alpha_2$, $|V_{21}|=1.21$. The units used throughout for Rabi frequencies and relaxation rates are nsec^{-1} ; the amplifier length is given in cm. The gains α_2 and α_3 are then given in $\text{cm}^{-1}\text{nsec}^{-1}$.

In Fig. 3.2 we show the level populations, and the nondiagonal density matrix elements as functions of the amplifier coordinate. Below saturation of the Stokes field, their values are determined completely by the pump laser, as given by Eq. (3.2a-d). The saturation of the Stokes field alters however strongly this behavior and tends to equalize

the populations of levels $|2\rangle$ and $|3\rangle$. In the small gain regime the population is mainly in states $|1\rangle$ and $|2\rangle$, but when the Stokes field becomes strong, it acts on the $|2\rangle \leftrightarrow |3\rangle$ transition like on a two level system, coupling these levels strongly and tending to equalize that populations.

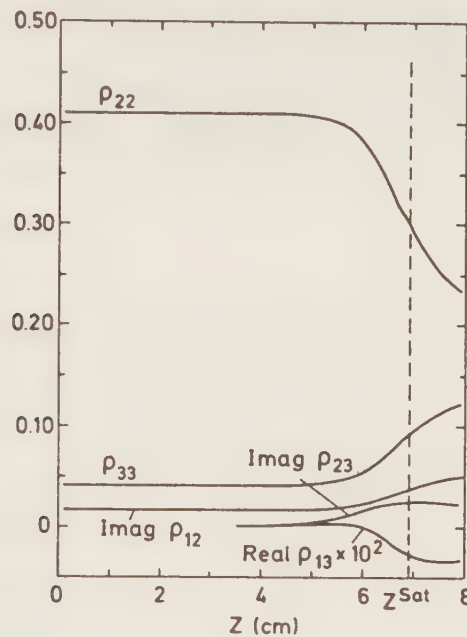


Fig. 3.2

Density matrix elements as function of the amplifier coordinate. The dashed line indicates the amplifier length z_{sat} at which the Stokes intensity reaches the saturation value I_2^{sat} . All parameters are as in Fig. 3.1. For clarity, we show only the density matrix elements relevant for the resonant amplification of the Stokes and idler fields.

3.4 Linear Stokes gain

In this Section we analyze in detail the Stokes gain in the linear regime (before saturation is reached), and its dependence on the various

system parameters. We start by the simplest case, where the Stark shifts are arbitrarily set equal to zero (*) and the pump field is applied at exactly the two-photon resonance ($\Delta_1=0$). (Note that switching off the Stark shifts produces a one to one correspondence between this system and the usual, λ -type three-level system /36-42/). Thus we have $\Delta_2=-\Delta_3$, and an exact analytical expression for the Stokes gain can be obtained:

$$\alpha'_2 = \frac{\alpha_2 \left(\frac{1}{\gamma_2} - \frac{1}{\gamma_3} \right) I_1^2}{2T_1 (1 + I_1^2)} \left(\frac{\frac{\gamma_{13}}{2} - \frac{\gamma_{13} - \gamma_{23}}{2} \frac{\Delta_2}{2a}}{(\Delta_2 - a)^2 + b^2} + \frac{\frac{\gamma_{13}}{2} + \frac{\gamma_{13} - \gamma_{23}}{2} \frac{\Delta_2}{2a}}{(\Delta_2 + a)^2 + b^2} \right), \quad (3.10a)$$

where

$$a^2 = |\Omega_{12}|^2 - \frac{(\gamma_{13} - \gamma_{23})^2}{4} \quad (3.10b)$$

$$b^2 = \frac{(\gamma_{13} + \gamma_{23})^2}{4}. \quad (3.10c)$$

The gain spectrum of Eq. (3.10a) is plotted in Fig. 3.3 as a function of the pump intensity and the Stokes probe detuning Δ_2 . For weak pump fields, such that $|V_{21}|^2 \ll \gamma_{23} \gamma_{13}$, the gain profile tends to the Lorentzian

$$\alpha'_2 = \frac{\alpha_2 \left(\frac{1}{\gamma_2} - \frac{1}{\gamma_3} \right) I_1^2}{2T_1 \gamma_{23} (1 + I_1^2)} \frac{\gamma_{23}^2}{\Delta_2^2 + \gamma_{23}^2}. \quad (3.11)$$

(*) The Stark shifts (Eq. (2.14)) depend on the dipole moments and on the specific level configuration; therefore, under some special parameters or level configurations one could have the case where $\Delta_1 + \omega_{31} I_1 \approx 0$. In general that is not the case.

with a half-width γ_{23} . (*)

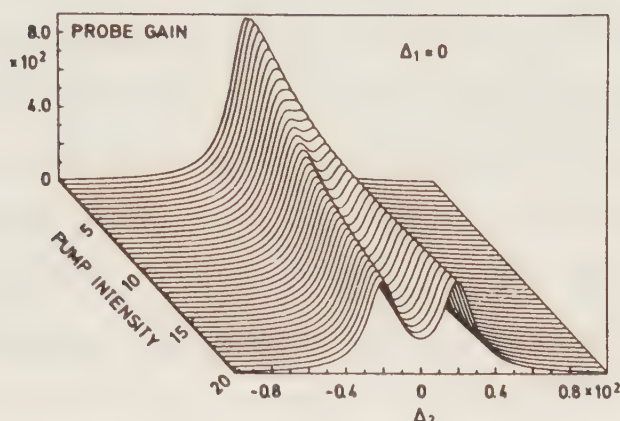


Fig. 3.3

Stokes probe gain spectrum as function of the pump intensity (in units of the Rabi frequency Ω_{21}). Here the Stark shifts are set equal to zero, and $\gamma_{12} = \gamma_{23} = \gamma_{13} = 10$, $\gamma_2 = 0.03$, $\gamma_3 = 1$.

As the pump intensity increases, the spectrum displays a double-peaked dynamic Stark splitting. This feature is characteristic of spectra obtained when probing a transition with a weak field coupled to a third level [48]. In our case, the upper-level of the two-photon transition is probed. The Stark splitting originates in the $D_2 D_3 |V_{21}|^2$ term in the denominator of α'_2 , Eq. (3.9c). One can see this more clearly by noting that for $\gamma_{13} \sim \gamma_{23}$ and large pump intensities, Eq. (3.10b) reduces approximately to $\alpha \sim \pm |\Omega_{21}|$, and the Stokes gain α'_2 can be reexpressed as the sum of two Lorentzians centred at $\pm |\Omega_{21}|$ (the two-photon Rabi frequency) and of half-width equal to the average value of the relaxation rates γ_{13} and γ_{23} :

(*) The two-photon saturation is governed by the factor $1 + I_1^2$ in the denominator of Eq. (3.10a). A non saturating field satisfies $|V_{21}|^2 \ll \gamma_2 / 4T_1$. This condition often requires weaker fields than those satisfying $|V_{21}|^2 \ll \gamma_{23} \gamma_{13}$, due to the long upper-state lifetime.

$$\alpha_2' = \frac{\alpha_2 \left(\frac{1}{\gamma_2} - \frac{1}{\gamma_3} \right) I_1^2 \gamma_{13}}{\tau_1 (1 + I_1^2) (\gamma_{13} + \gamma_{23})^2} \left[\frac{(\gamma_{13} + \gamma_{23})^2 / 4}{(\Delta_2 \pm |\Omega_{21}|)^2 + (\gamma_{13} + \gamma_{23})^2 / 4} \right]. \quad (3.12)$$

Fig. 3.4 shows the Stokes gain profiles for a detuned pump (here $\Delta_1 = -10$), the Stark shifts still being set equal to zero. The different curves correspond to various pump intensities, from 2.5 to 20 by steps of 2.5 in units of the Rabi frequency $|\Omega_{21}|$. As expected, the Stark splitting increases with the pump field intensity. In contrast to the case of resonant pumping ($\Delta_1 = 0$), the spectrum is now asymmetric. This is further illustrated in Fig. (3.5), where we plot the Stokes gain profile as a function of both detunings Δ_1 and Δ_2 for a fixed pump intensity. At the two-photon resonance, the profile shows two peaks equidistant from $\Delta_2 = 0$. Pumping far from resonance yields one peak located almost at $\Delta_2 = 0$, and a second at $\Delta_2 = \Delta_1$, which corresponds to the Raman line.

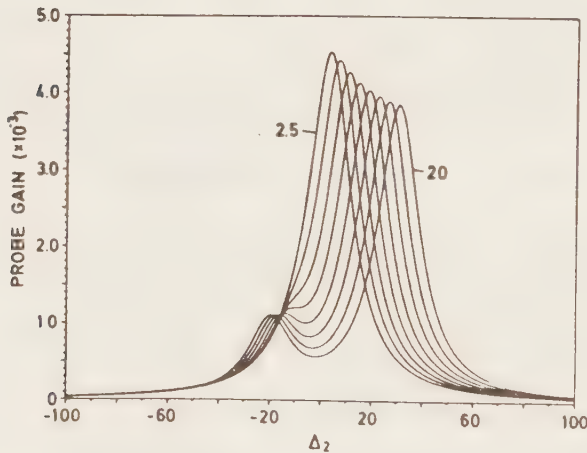


Fig. 3.4

Probe gain as a function of the probe detuning Δ_2 for various values of the pump intensity, and $\Delta_1 = -10$. The curves are monotonic in intensity, so that only the outer two are labeled. Step in intensity 2.5 (in units of the Rabi frequency $|\Omega_{21}|$). Relaxation rates as in Fig. 3.3.

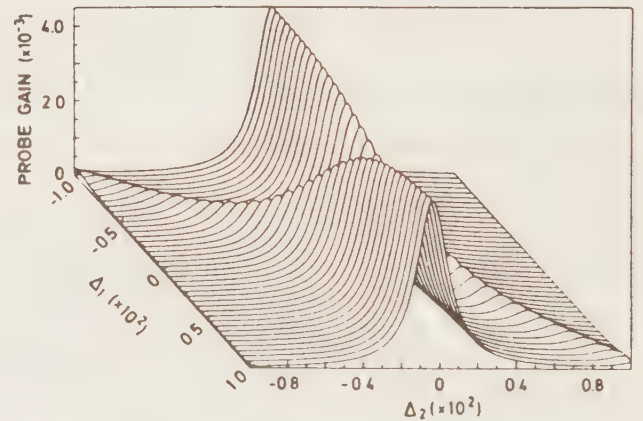


Fig. 3.5

Probe gain as function of Δ_1 and Δ_2 , for a fixed pump intensity $\gamma_{21} = 20$. Relaxation rates as in Fig. 3.3.

We now discuss the effect of Stark shifts on the probe gain profiles. We consider first the Stark shifted resonance, i.e., $\Delta_1 = -\omega_s I_1$. The effective pump detuning is $\Delta_1' = \Delta_1 + \omega_s I_1 = 0$, so that we obtain symmetric curves like those of Fig. 3.3 but shifted by $\omega_s I_1$. The gain spectrum at the real two-photon resonance $\Delta_1 = 0$ as a function of the pump intensity with the Stark shifts is shown in Fig. 3.6. The gain profiles are asymmetric as those of Fig. 3.4, and clearly show the effect of Stark shifts as pump intensity dependent detunings.

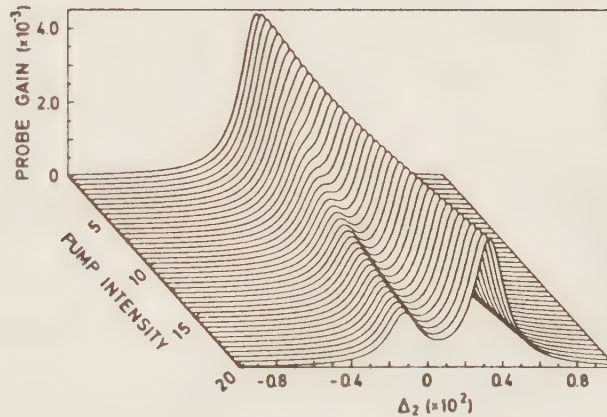


Fig. 3.6

Effect of the Stark shifts on the probe gain as function of Δ_2 and pump intensity (in units of the Rabi frequency Ω_{21}). $\Delta_1 = 0$ and $\omega_{s1} = \omega_{s2} = -\sqrt{\gamma_2 \gamma_{21}}/4$. Relaxation rates as in Fig. 3.3.

A similar behavior of the Stark splitting was observed experimentally in the ordinary three-level system /48/, except that the splitting is determined by the usual pump Rabi frequency, instead of the effective two-photon Rabi frequency as in our case. Experiments in two-photon pumped systems were performed by Heinrich /12/ and by Cotter et al. /10/, who measured the Stokes intensity as a function of pump detuning in Cd and Na vapor, respectively. They found a dip at the two-photon resonance, and a line width 20 to 30 times larger than both the

linewidth of the pump laser and the Doppler width of the $|2\rangle \leftrightarrow |3\rangle$ transition. The dip was tentatively attributed to a strong two-photon absorption or three-photon ionization. We offer here an alternative explanation based on the effects of Stark splitting and power broadening. In Fig. 3.7 we show the Stokes gain as a function of the pump detuning Δ_1 and probe detuning Δ_2 . Close to the atomic transition $|2\rangle \leftrightarrow |3\rangle$ ($\Delta_2=0$), its spectrum exhibits a dip at the two-photon resonance ($\Delta_1=0$), and is strongly broadened. This broadening is the power broadening arising from the term $\omega_{s1}I_1$ in D_1 (see Eq. (3.1a)).

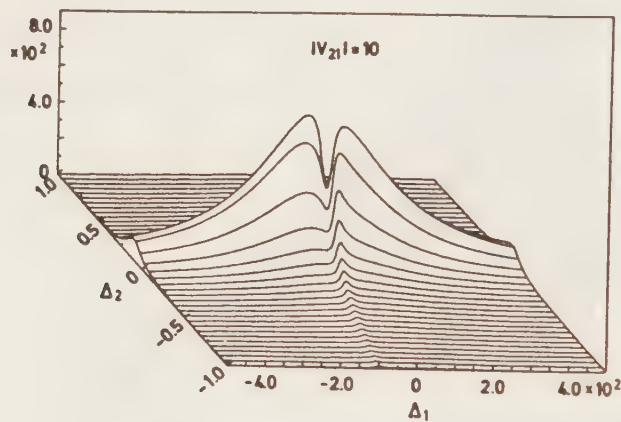


Fig. 3.7

Probe gain as function of the pump and probe detunings Δ_1 and Δ_2 for a fixed pump intensity. $|V_{21}|=10$, $V_2=0.1$, $V_3=1$, $V_{21}=V_{13}=V_{23}=10$.

In Fig. 3.8 and 3.9 we compare the experimental data from ref. /12/ with our theoretical results. The line width of the Stokes spectrum for fixed pump frequency (Fig. 3.8) is determined by the decay rates, the field being too weak to give a Stark splitting. Nevertheless, a small dip in the Stokes intensity as a function of pump detuning can be observed (Fig. 3.9); it corresponds to the situation of Fig. 3.7, and practically disappears at low intensities. This profile is power broadened.

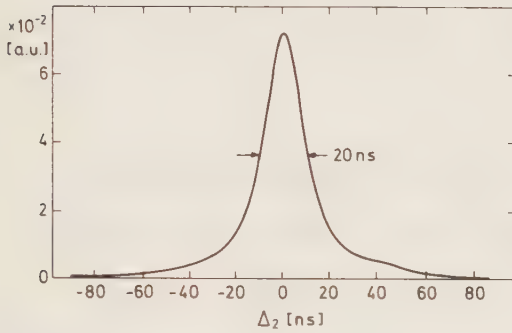
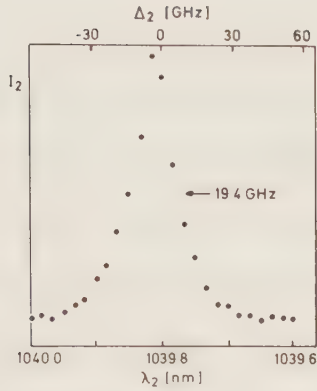


Fig. 3.8

Comparison between the measured Stokes spectrum of ref./12/ (up) and a theoretical probe gain profile (down). Relaxation rates as in Fig. 3.7.

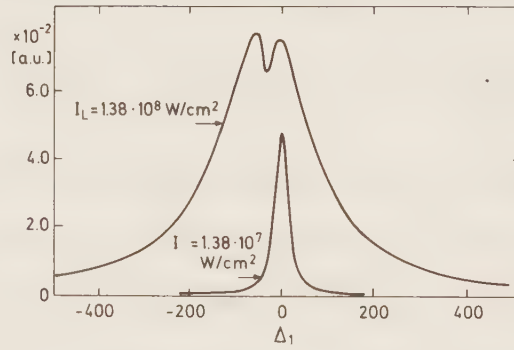
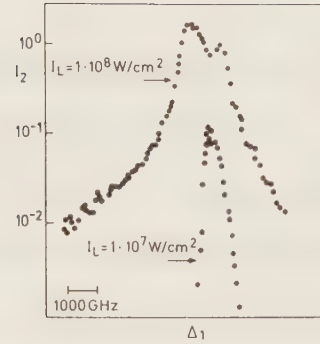


Fig. 3.9

Up: Measured Stokes intensity as a function of the pump detuning. Down: Theoretical probe gain profile, calculated with the same relaxation rates as in Fig. 3.7.

We note however that an exact, quantitative comparison between our steady-state, weak probe calculation and the experimental results is not possible; pulse propagation through the medium leads to a broadening of the Stokes spectrum (pulse shortening), and this effect is obviously not included here. Indeed, a quantitative comparison shows that the experimental Stokes intensity spectra reproduced here are broader than the computed ones. Nonetheless, our model gives a simple physical interpretation of the spectral shape of the Stokes line.

3.5 Saturation

Until now, we have considered the behaviour of the system at weak probe and idler intensities. For large atomic densities (10^{16} cm^{-3}) however, the Stokes gain is large and the exponential growth law of the Stokes line rapidly ceases to be valid. In this Section, we give an approximate, analytical description of the saturation mechanism leading to a departure from exponential growth.

3.5.1. Saturation of the Stokes field

In order to gain some analytical insight, we assume here that the Stokes field $\mathcal{E}_2$ reaches saturation before the idler. That is, we assume that the idler field $\mathcal{E}_3$ does not affect strongly $\mathcal{E}_2$, so that we can neglect the Stokes-idler coupling for $\mathcal{E}_2$ but not for $\mathcal{E}_3$. At the end of this Section, this assumption will be checked against numerical results and shown to be well satisfied. We can then study the onset of saturation by solving the density matrix equations to second order in the Stokes field. The relevant coherence for the evolution of $\mathcal{E}_2$ is ρ_{23} , which is found to be

$$\rho_{23} = \frac{i V_{23} S_{23}^*}{S_1 S_2} (D_2 \beta + D_1^* D_2 D_3 |V_{21}|^2) . \quad (3.13)$$

where the Stokes saturation denominator S_2 is given by

$$S_2 = |S_{23}|^2 + \frac{|V_{23}|^2}{S_1} \frac{2\mathcal{L}_2}{\gamma_2 \gamma_{23}} \left[1 + \frac{3I_1^2 \mathcal{L}_1}{2\gamma_3 T_1} \left(1 + \frac{\mathcal{L}_3 |V_{21}|^2}{\gamma_{13} \gamma_{23}} \right) \right] . \quad (3.14)$$

We have introduced the Lorentzians

$$\mathcal{L}_2 = \gamma_{23}^2 / (\gamma_{23}^2 + (\Delta_2 + \omega_{s2} I_1)^2)$$

and

$$\mathcal{L}_3 = \gamma_{13}^2 / (\gamma_{13}^2 + (\Delta_3 + \omega_{s3} I_1)^2)$$

When all quadratic terms are neglected, ρ_{23} reduces to the linear regime solution (Eq. (3.2e)), with $V_{13} = 0$, i.e., negligible Stokes-idler coupling. Substituting Eq. (3.13) into the field equation (2.27b) gives the evolution of the Stokes intensity:

$$\frac{dI_2}{dz} = \frac{\mathcal{G} I_2}{1 + \frac{I_2}{I_2^{\text{sat}}}} \quad (3.15)$$

where

$$\mathcal{G} = \frac{2\alpha_2 \beta \mathcal{L}_2}{\gamma_{23} S_1 |S_{23}|^2} \left(1 + \frac{\mathcal{L}_2}{\gamma_{23} \gamma_{13}} |V_{21}|^2 \right) \quad (3.15a)$$

The saturation intensity I_2^{sat} , defined as usual as the intensity at which the gain is reduced to half the linear gain, is given by :

$$I_2^{\text{sat}} = \frac{\gamma_2 \gamma_{23} S_1 |S_{23}|^2}{\left(\frac{d_{23}}{2\hbar} \right)^2 \mathcal{L}_2 \left[1 + \frac{3I_1^2 \mathcal{L}_1}{2\gamma_3 T_1} \left(1 + \frac{\mathcal{L}_3 |V_{21}|^2}{\gamma_{13} \gamma_{23}} \right) \right]} \quad (3.16)$$

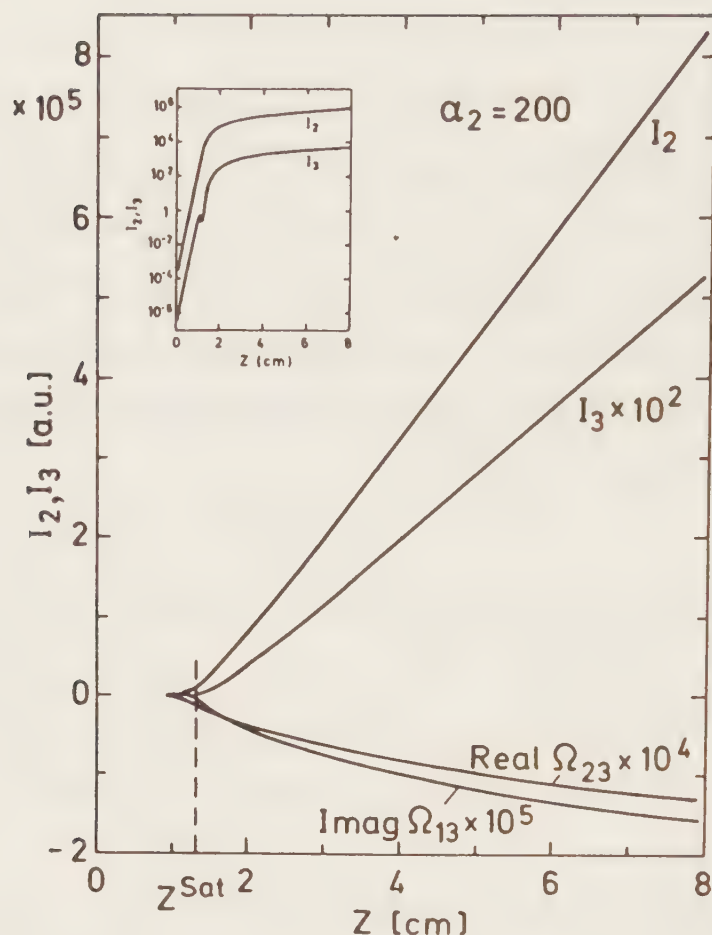


Fig. 3.10

Rabi frequencies and field intensities (in arbitrary units) inside the saturation regime as function of the amplifier coordinate in a linear scale. The inset shows the intensities in a log-scale. Relaxation rates and pump intensity as in Fig. 3.1.

The value I_2^{sat} in Fig. 3.1 has been calculated from this expression. Fig. 3.10 shows the numerical solution for the Rabi frequencies and the field intensities as functions of z when saturation sets in. In Fig. 3.11 we show the corresponding populations and nondiagonal matrix elements. All Stark shifts and detunings have been set to zero. For values $I_2 \gg I_2^{\text{sat}}$ the Stokes field intensity grows linearly with the

amplifier coordinate, as usual. Such behaviour can easily be understood by inspection of Eq. (3.15). For $I_2/I_2^{\text{sat}} \gg 1$, we can neglect the one in the denominator and obtain:

$$\frac{dI_2}{dz} \approx \ell I_2^{\text{sat}}. \quad (3.17)$$

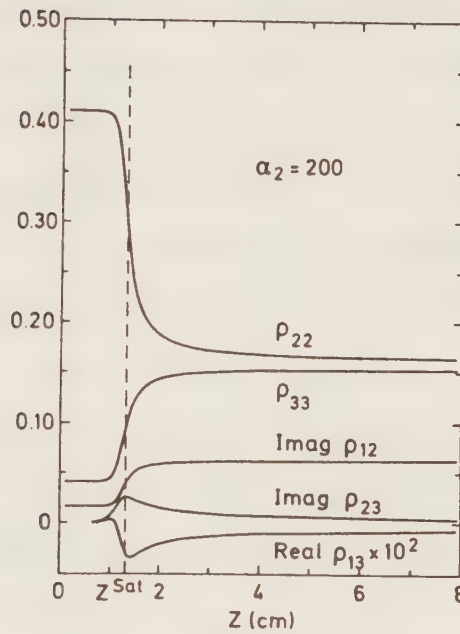


Fig. 3.11

Density matrix elements inside the saturation regime as a function of the amplifier coordinate. Relaxation rates and pump intensity as in Fig. 3.1.

which yields immediately $I_2 \sim I_2^{\text{sat}} \ell z$. We have compared this analytical result with an exact numerical solution and found that ℓ differs from the exact value by about 10%. This indicates that the coupling with the idler field does not strongly affect the propagation of the Stokes field, and justifies a posteriori our approximate analytical approach.

3.5.2 Saturation of the idler field

Contrary to the Stokes line, the idler-Stokes coupling terms cannot be neglected when studying the propagation of the idler field. This is because the growth of the idler line is strongly coupled to that of the Stokes line, not only in the small signal regime but also in the saturation regime. In this case the analytical solutions become extremely cumbersome, and one has to rely on numerical solutions. The results of this analysis are summarized in Figures 3.10 and 3.11 for the case of resonant two-photon pumping. In the saturation regime, the idler intensity, as the Stokes intensity, grows linearly along the medium. The density matrix elements tend to constant values, and populations in the states $|2\rangle$ and $|3\rangle$ tend to equalize. The effect of phase mismatch can be neglected, provided that the phase mismatch Δk is much smaller than α_2 . In this case, it simply introduces an extra phase to the idler field, so that neither oscillations nor a large attenuation are observed in the intensity (see Fig. 3.12). For large Δk , however, as would be the case e.g. when working with higher excited states, the phase mismatch not only can cause intensity oscillations, but also strongly reduces the output intensity.

The effect of the Stark shift is more important. It acts as an intensity dependent detuning, introducing phase shifts into the fields, and diminishes the intensities (Fig. 3.12). The mixing of the Rabi frequencies also blurs the dip in the idler intensity near the onset of the saturation regime.

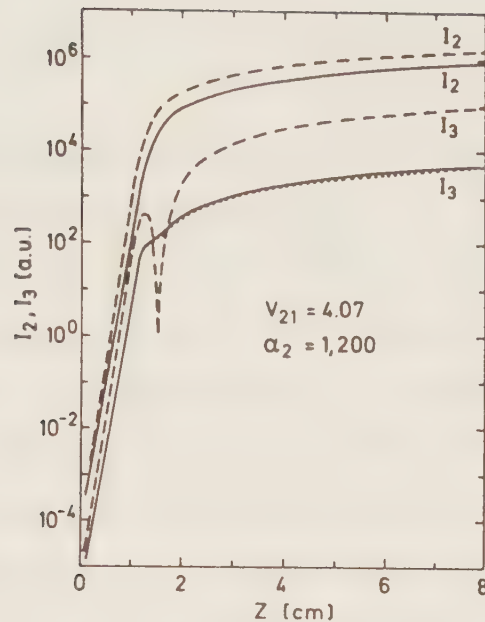


Fig. 3.12

The effect of phase mismatch and Stark shifts on the Stokes and idler intensities as function of the amplifier coordinate z at $\Delta_1 = 0$. Dashed lines: without Stark shifts and phase mismatch. Full lines: with Stark shifts. Dotted line: with Stark shifts and phase mismatch. Stark shifts and phase mismatch values are those calculated for Cd.

Saturation curves for the idler field as a function of the atomic density have been measured in Sodium [16]. While the growth of the fields is studied experimentally by varying the active atomic density (N) at constant interaction length, the theoretical results are given in terms of the amplifier coordinate (z) at fixed atomic density. In the linear regime as well as in the saturation regime, the field shows the same functional dependence on the atomic density as on the amplifier coordinate, so that both type of curves are equivalent. (*) The

(*) It is sometimes useful to introduce the variable (Nz) , which accounts for variations both in z and in N .

experimental curves /16/ are in qualitative agreement with our results.

3.6 Comparison with Cadmium experiments

Fig. 3.13 shows the experimental dependence of the Stokes intensity on the atomic density /12/. For the high atomic densities used here ($N=10^{17}-10^{18} \text{ cm}^{-3}$), the system is well inside the saturation regime, and the field intensity grows linearly with the atomic density and/or the amplifier length. It is important to realize that this behavior is due to the saturation of the transition $|2\rangle \leftrightarrow |3\rangle$ caused by the Stokes field. As we have seen, first-order perturbation theory in the Stokes field would predict an exponential growth of I_2 , even when the two-photon transition is saturated (see Eqs. (3.7a) and (3.4d)).

In Fig. 3.14 we have plotted the theoretical Stokes and idler intensities as functions of the pump intensity and compared our results with the experimental data /12/. The theoretical curves were calculated for an atomic density two orders of magnitude smaller than the experimental ones in order to keep the computing time within reasonable bounds. The qualitative agreement is however quite good. The complicated functional dependence of I_2 and I_3 on the pump intensity reflects the complicated dependences of I_2^{sat} and the slope $\mathcal{C}$ on I_L through V_2 , and I_1 (Eq. 3.15a and 3.16).

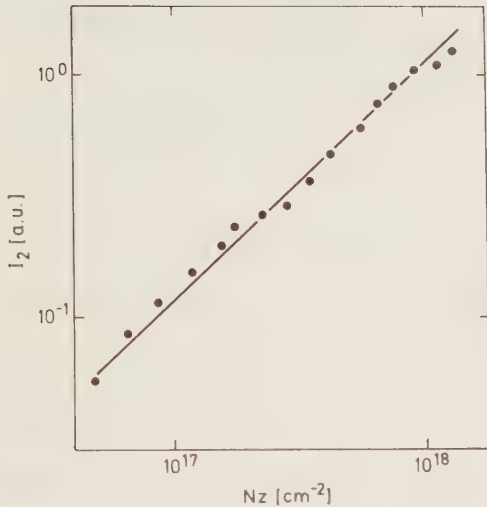


Fig. 3.13

Stokes intensity (in arbitrary units) as a function of N_z . Comparison between the measurements of ref. /12/ (dots) and our theoretical results (full curve). ($I_L = 3 \times 10^7 \text{ W/cm}^2$).

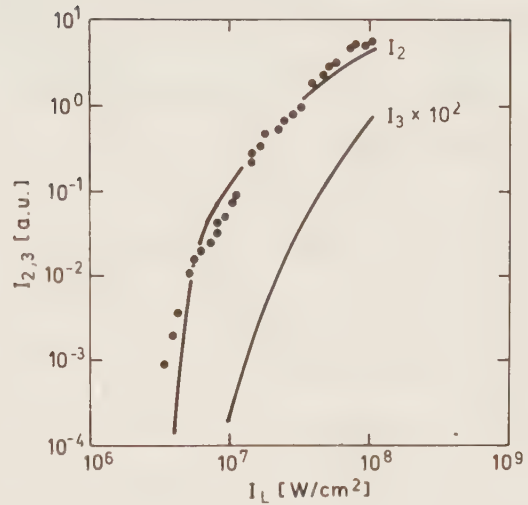


Fig. 3.14

Stokes (I_2) and idler (I_3) intensities (in arbitrary units) as function of the pump intensity. The dots correspond to the measurement of ref. /12/. ($N_{z_{exp}} = 2 \times 10^{17} \text{ cm}^{-3}$).

The calculations were performed for resonant pumping, and including Stark shifts. We have considered the effect of phase mismatch for the case of weakly focused pump beams, where the plane wave assumption can be done. It has been however realized /6/ that phase mismatch effects can be very different when working with tight focusing. Nevertheless the agreement with experimental results is good, which possibly indicates that the effect of phase mismatch can be neglected in the forward direction.

The Stokes field is not tunable: its frequency remains fixed as the pump detuning is varied /12/. This can be understood from Fig. 3.5; when pumping near resonance ($\Delta_f = 0$), the Stokes field is generated mostly by

lasing from the upper state $|2\rangle$, as evidenced by the Lorentzian profile of the probe gain about $\Delta_2=0$, which is much stronger than the tunable 'hyper-Raman line', having its maximum around $\Delta_2=\Delta_1$. Nevertheless SHRS can be observed for large pump detunings (Fig. 3.9) due to the large power broadening.

CHAPTER IV

TRANSIENT REGIME. - SQUARE PUMP PULSES.

4.1 Introduction

The study of transients in three-level systems has been the object of considerable work (see for example /31,36,41, 45,53-60/), dealing in particular with effects such as optical nutation /53/ and free induction decay /54-56/, as well as with other more elaborate techniques including photon echoes /53-54,57-58/, delayed pulse spectroscopy /54/, self-induced transparency /59/, two-pulse delayed spectroscopy /54/, etc.. The measurement of such transients provides information on atomic or molecular systems complementary to that given by the methods of saturation spectroscopy: while the last ones yield information in the frequency domain, transient effects give directly time domain information and as such constitute a powerful tool for measuring relaxation rates. It is natural to look for spectroscopic techniques combining the advantages of both techniques, i.e., which deliver simultaneously time and frequency domain information. Feld /60/ and Ducloy et al./31/ proposed such a technique. It consists in applying a weak probe field to the system, and measuring the gain profiles after various delay times, or equivalently, measuring the transient behaviour at various probe frequencies.

A similar technique can be applied to study the dynamical behavior of two-photon pumped systems. In this chapter, we discuss the situation where a weak, tunable field $\mathcal{E}_2$ near the atomic frequency ω_{23} probes the $|2\rangle \leftrightarrow |3\rangle$ transition, while a strong field $\mathcal{E}_1$, pumping the $|1\rangle \leftrightarrow |2\rangle$ transition is suddenly switched on or off (See Fig. 4.1a). The effects

associated with the switching on of $\mathcal{E}_1$ at time $t=0$ (Fig. 4.1b) are usually referred to as optical nutation effects, and are discussed specifically in Section 4.4, while the free-induction decay type effects associated with the switch-off of $\mathcal{E}_1$ at $t=t_0$ are studied in Section 4.5. The experimental arrangement discussed in this Chapter is of course not the only possible one: for instance one could also probe the $|3\rangle \leftrightarrow |1\rangle$ transition, in order to study idler dynamics, or use two probes.

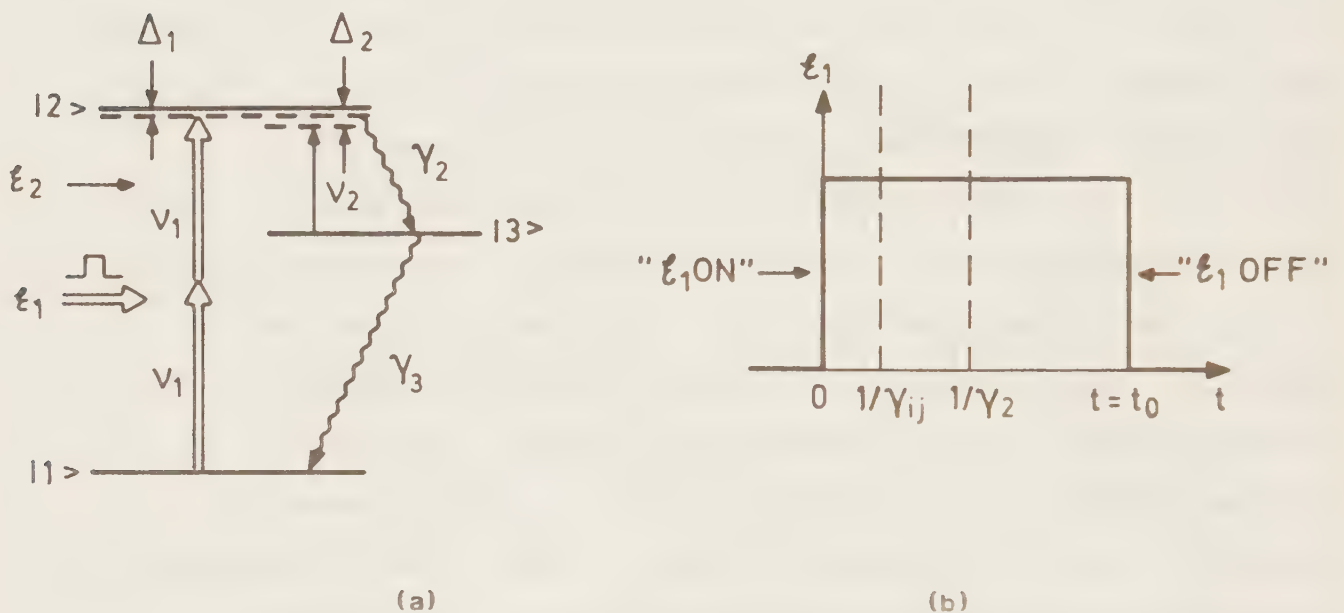


Fig. 4.1

(a) Level scheme with the weak c.w. probe $\mathcal{E}_2$ at frequency ν_2 and the strong square pump pulse of frequency ν_1 . (b) Square pump pulse longer than the inverse of phase and upper state population relaxation rates γ_{21} , and γ_2 . Optical nutation is observed after the field is switched on ($t>0$), and free induction decay after the pump is switched off ($t>t_0$).

We note that transient, heterodyne detection schemes such as optical nutation experiments yield information not obtainable by frequency domain measurements. In particular, we will see that they provide, e.g., the phase relaxation rate γ_{21} of the two-photon coherence. This,

however, is obviously not measurable via standard linewidth methods, since the $|1\rangle \leftrightarrow |2\rangle$ transition is dipole forbidden in our case.

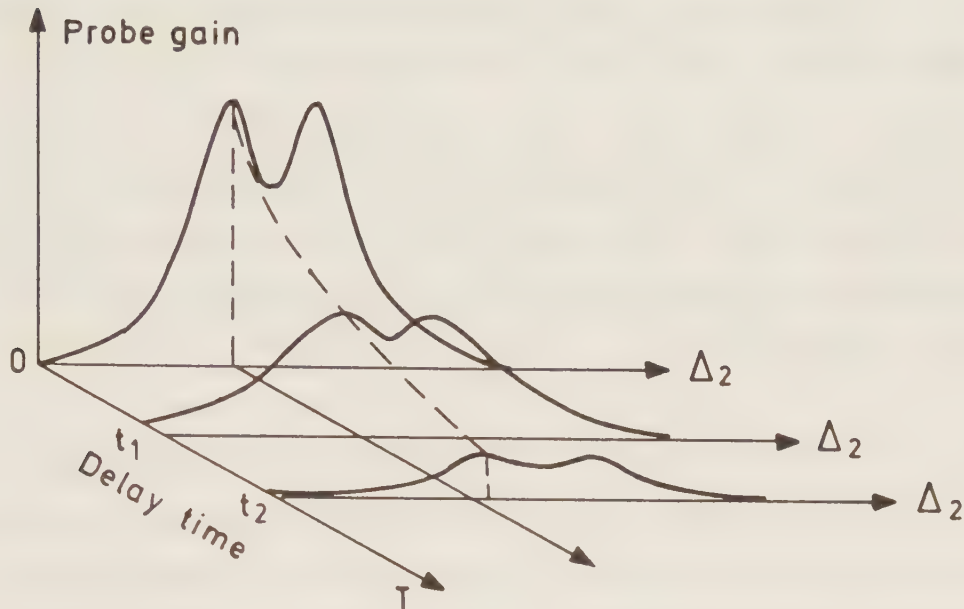


Fig 4.2

Illustration of a delayed probe gain spectrum. The various spectral shapes at fixed times give information about transient frequencies, while delayed gain measurements at fixed probe frequencies give information about the relaxation processes.

A typical time-dependent probe gain measurement yields raw data as schematically illustrated in Fig. 4.2 where the probe gain is shown as a function of both probe detuning Δ_2 and delay time after switching the pump field on or off. The temporal decay of the gain yields information on relaxation rates, and if it is oscillatory, its period of oscillation gives information on the Rabi frequencies. From the gain profile, one can also learn about the transient frequencies, Stark shifts and Stark splittings. The method is also a valuable tool to differentiate between Raman- and population-type processes.

The major flaw of pump-probe techniques, when applied to complicated systems, is that the results typically depend on a considerable number of parameters. In order to simplify the analysis of the raw data, it is therefore crucial to always work -both experimentally and theoretically- in limiting cases where at least some of the parameters can be eliminated. General solutions are in fact extremely complicated, and that is why we consider in this chapter only some limiting cases. Our choice is somewhat arbitrary, and is meant for illustrating the type of information which can be gained from pump-probe measurements in two-photon pumped systems.

In all cases, the method applied to find the gain spectrum is to solve the density matrix equations (2.11) perturbatively to first order in the probe field. We work here within the thin sample approximation, which is summarized in the next Section. This method is almost always used in spectroscopy, since it avoids complications due to propagation and phase-matching effects and thus allows an easier analysis of the data. Our transient analysis is valid for adiabatic square pump pulses, i.e., pulses which rise in a time short compared with the inverse of the relaxation rates but slowly enough to satisfy the adiabatic criterion (A1.8) with respect to the intermediate state detunings.

4.2 The thin sample approximation

By 'optically thin sample', one means an active medium of length L such that $gL \ll 1$, where g is the gain for an externally applied field. One can understand this approximation quantitatively by formally solving

the field equation (2.22) (*). This gives:

$$E(z, t) = E(0, t - z/v) + ik/2 \epsilon_0 \int_0^z dz' E(z', t - (z - z')/v). \quad (4.1)$$

The first term on the right hand side of Eq. (4.1) describes the free propagation of the incident field, while the integral is the radiated field and describes the change in E as it propagates through the medium. For an optically thin sample of length L , we assume that the radiated field remains small compared to the free field contribution. Writing the total field in Eq. (4.1) as $E(z, t) = E_F(z, t) + E_R(z, t)$, where E_F is the free and E_R the radiated field, we can approximate $E(z, t)$ by $E(0, t - z/v)$ when calculating the polarization. This is the optically thin sample approximation. In general, one obtains that $P = \epsilon_0 N \chi(E) E$, where $\chi(E)$ is the nonlinear susceptibility of the medium. For weak probe fields, $\chi(E)$ is independent of the probe intensity and we are able to write

$$P(z, t) = \epsilon_0 N \chi E_F(0, t - z/v) \quad (4.2)$$

With the approximate form (4.2), Eq. (4.1) can readily be integrated to give

$$E(L, t) = E(0, t - L/v) (1 + gL). \quad (4.3)$$

where the complex small field gain g is given by $g = ikL\chi/2$. Note also that consistently with this approximation we can neglect phase-matching effects.

(*) We neglect background absorption.

The total intensity detected at the end of the sample is proportional to $|E(L)|^2 = |E_F + E_R|^2$, which becomes, for a weak radiated field

$$|E(L)|^2 \simeq |E_F|^2 + 2\text{Re}(E_R^* E_F) \quad (4.4)$$

It is the second term in Eq. (4.4) which yields information about the optical properties of the medium. It is a beat signal between the free and radiated fields, and can experimentally be separated from the first one by heterodyning techniques.

4.3 Density matrix equations in the weak probe limit.

As already discussed, we obtain an analytical expression for the probe gain by solving the density matrix equations to first order in the probe field, assuming $V_{13} = 0$ and treating the pump field exactly. Explicitely, inserting the zeroth-order solution in V_{23} of the density matrix equations (2.11)

$$\begin{aligned} \dot{\rho}_{11}^{(0)} &= \gamma_3 \rho_{33}^{(0)} + (iV_{21} \rho_{12}^{(0)} + \text{c.c.}) \\ \dot{\rho}_{22}^{(0)} &= -\gamma_2 \rho_{22}^{(0)} - (iV_{21} \rho_{12}^{(0)} + \text{c.c.}) \\ \dot{\rho}_{33}^{(0)} &= \gamma_2 \rho_{22}^{(0)} - \gamma_3 \rho_{33}^{(0)} \\ \dot{\rho}_{21}^{(0)} &= -(\gamma_{21} + i\Delta_1) \rho_{21}^{(0)} + iV_{21} (\rho_{22}^{(0)} - \rho_{11}^{(0)}) \\ \rho_{13}^{(0)} &= 0 \\ \rho_{23}^{(0)} &= 0 . \end{aligned} \quad (4.5)$$

into the equations for the coherences ρ_{23} and ρ_{13} yields the first order

corrections

$$\begin{aligned}\dot{\rho}_{23}^{(1)} &= -(\gamma_{23} + i\Delta_2) \rho_{23}^{(1)} + iV_{23}(\rho_{22}^{(0)} - \rho_{33}^{(0)}) - iV_{21}\rho_{13}^{(1)} \\ \dot{\rho}_{13}^{(1)} &= -(\gamma_{13} - i\Delta_3) \rho_{13}^{(1)} + i\rho_{12}^{(0)}V_{23} - iV_{12}\rho_{23}^{(1)}.\end{aligned}\quad (4.6)$$

Here, the subscript denotes the order of perturbation in V_{23} . Assuming the relaxation rate γ_3 of the population of level $|3\rangle$ to be much faster than γ_2 , we obtain approximate analytical solutions of Eq. (4.5) and (4.6) for some particular cases by means of Laplace transform. The condition, $\gamma_3 \gg \gamma_2$, is assumed to be satisfied in the following sections.

4.4 Optical Nutation

Here we assume that the atomic system is initially in its ground state with the c.w. weak probe $\mathcal{E}_2$ on. The strong pump field $\mathcal{E}_1$ is then switched on suddenly at time $t=0$ (see Fig. 4.1b). From the previous Chapter we know that in steady-state, the gain spectrum consists of two peaks which may be asymmetric because of Stark shifts. In the initial atomic state there are however neither Stark splitting, nor Stark shifts: the evolution of the gain spectrum for $t>0$ gives therefore information on the onset of Stark shifts and splittings as well as on effective Rabi frequencies and decay rate constants.

Because the exact analytical solution of Eq. (4.5) and (4.6) is very complicated, we limit our discussion to two especial cases: in the first one, the pump field is assumed to be at resonance with the Stark shifted $|1\rangle \leftrightarrow |2\rangle$ transition ($\Delta_1^* = \Delta_1 + \omega_{S1} = 0$), while in the second case, it is assumed to be so strong that a perturbative treatment in the relaxation rates can be made.

4.4.1. Pumping at the Stark shifted resonance $\Delta_1' = 0$.

At the Stark shifted resonance $\Delta_1' = 0$, conservation of energy implies that $\Delta_2' = -\Delta_3'$. Introducing these values of the detunings in Eqs. (4.4) and (4.5) and Laplace transforming them gives

$$\bar{\rho}_{22}^{(0)}(s) = \frac{2|V_{21}|^2 (s + \gamma_3)}{s D_{21}(s)} \quad (4.7a)$$

$$\bar{\rho}_{21}^{(0)}(s) = \frac{-i V_{21} (s + \gamma_2) (s + \gamma_3)}{s D_{21}(s)} \quad (4.7b)$$

$$\bar{\rho}_{23}^{(1)}(s) = \frac{i V_{23}}{D_{23}(s)} \left\{ \left(1 - \frac{\gamma_2}{s + \gamma_3} \right) \bar{\rho}_{22}^{(0)}(s + \gamma_{13} - i \Delta_3') - i \bar{\rho}_{12}^{(0)} V_{21} \right\} \quad (4.7c)$$

where the initial conditions are $\rho_{11}(0) = 1$, and all other $\rho_{ij}(0) = 0$. $\rho_{23}^{(1)}$ gives the probe gain about the $|2\rangle \leftrightarrow |3\rangle$ transition, therefore we have kept only the density matrix equations relevant for its determination. The denominators $D_{21}(s)$ and $D_{23}(s)$ are given by

$$D_{21}(s) = (s + \gamma_3) (s + \gamma_{21}) (s + \gamma_2) + 2|V_{21}|^2 (2(s + \gamma_3) + \gamma_2) \quad (4.8a)$$

and

$$D_{23}(s) = (s + \gamma_{23} + i \Delta_2') (s + \gamma_{13} + i \Delta_2') + |V_{21}|^2 \quad (4.8b)$$

respectively. The roots of these polynomials are the poles of $\bar{\rho}_{23}^{(1)}(s)$, and determine the transient behavior of the system. Specifically, one finds six poles for the Laplace transformed gain in Eq. (4.7). Three of them are obtained straightforward: $s_1 = 0$, which gives the steady-state solution, and s_5, s_6 which are the roots of the second-order polynomial $D_{23}(s)$. The three roots of the polynomial $D_{21}(s)$ can be obtained perturbatively to first order in γ_2 , by applying the condition $\gamma_3 \gg \gamma_2$.

In order to do that we rewrite $D_{21}(s)$ as

$$D_{21}(s) = (s + \gamma_2) \{ (s + \gamma_2)(s + \gamma_{21}) + 4|V_{21}|^2 \} + 2|V_{21}|^2 \gamma_2.$$

and solve perturbatively in the last term. To zeroth-order we have

$$s_2^{(0)} = -\gamma_3$$

$$s_{3/4}^{(0)} = -\frac{\gamma_2 + \gamma_{21}}{2} \pm i \sqrt{4|V_{21}|^2 - \left(\frac{\gamma_2 - \gamma_{21}}{2}\right)^2}. \quad (4.9)$$

A first order approximation to s_2 , s_3 and s_4 can be obtained by using the technique of Newton's expansion (see e.g. /61/), which is usually applied to find the roots of transcendental equations and gives good convergence as long as the derivative of the function at the initial probe value is not nearly zero:

$$s_i^{(1)} = s_i^{(0)} - \bar{P}_{23}^{(1)}(s_i^{(0)}) / \frac{d\bar{P}_{23}^{(1)}}{ds}(s_i^{(0)}) \quad (4.10)$$

The six roots are then approximately given by

$$s_1 = 0$$

$$s_2^{(1)} = -\gamma_3 - \frac{\gamma_2}{2} A$$

$$s_{3/4}^{(1)} = -\frac{\gamma_2 + \gamma_{21}}{2} + \frac{\gamma_2}{4} A \pm i \sqrt{4|V_{21}|^2 - \left(\frac{\gamma_2 - \gamma_{21}}{2}\right)^2} \left\{ 1 + \frac{\gamma_2}{4} A \frac{\gamma_3 - \frac{\gamma_2 + \gamma_{21}}{2}}{4|V_{21}|^2 - \left(\frac{\gamma_2 - \gamma_{21}}{2}\right)^2} \right\}$$

$$s_{5/6} = -\frac{\gamma_{13} + \gamma_{23}}{2} - i \Delta_2 \pm \frac{i}{2} \sqrt{4|V_{21}|^2 - (\gamma_{23} - \gamma_{13})^2}. \quad (4.11)$$

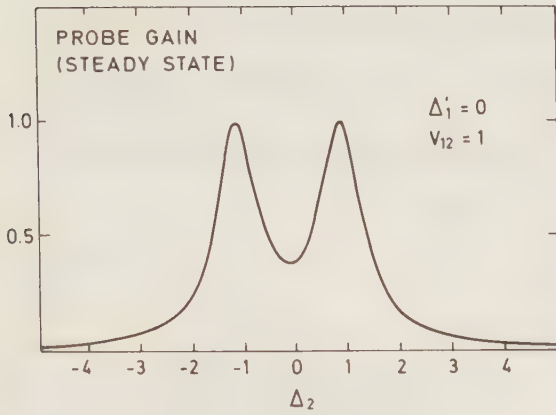
where $A = 4|V_{21}|^2 / (4|V_{21}|^2 + (\gamma_2 - \gamma_3)(\gamma_{21} - \gamma_3))$.

From the Stokes field equation (2.27b), we obtain that the probe gain profile is given by the imaginary part of the inverse Laplace transform of $\bar{\rho}_{23}^{(1)}(s)$. This reduces to the sum of the residues of the different poles, and has the form:

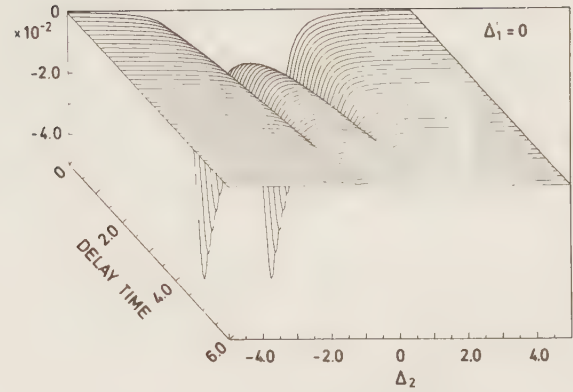
$$\rho_{23}^{(1)}(t) = \sum_i \text{Res}_i \{ \bar{\rho}_{23}^{(1)}(s) \} \exp(s_i t) \quad (4.12)$$

We study here each contribution to $\rho_{23}^{(1)}(t)$ separately:

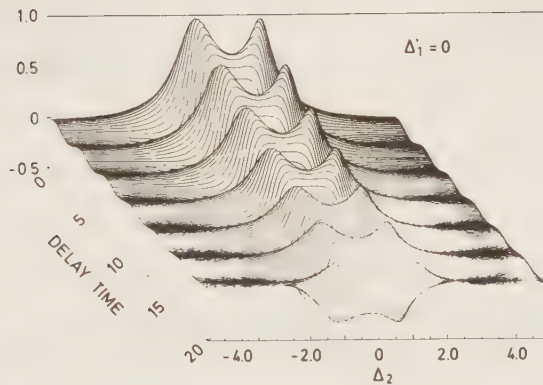
- The steady-state contribution ($s_1 = 0$) is the same as obtained in the preceding chapter, and can be seen in Fig. 4.3a. The gain spectrum consists of two peaks centred at $\Delta_2 = \pm |V_{21}|$.
- The s_2 -contribution (Fig. 4.3b) also exhibits two peaks centred at $\Delta_2 = \pm |V_{21}|$, but is negative. This transient decays essentially with the population relaxation rate γ_3 of state $|3\rangle$.
- Depending upon the relative magnitudes of $|V_{21}|$ and $|\gamma_2 - \gamma_{21}|/4$, s_3 and s_4 can give either rise to pure exponential decay or to damped oscillations. Two situations are of particular spectroscopic relevance. In the weak field limit, $|V_{21}| \ll |\gamma_2 - \gamma_{21}|/4$, the roots s_3 and s_4 yield exponential decays at rates γ_2 and γ_{21} . This regime is therefore appropriate to measure atomic decay rates. In the opposite limit, $|V_{21}| \gg |\gamma_2 - \gamma_{21}|/4$, s_3 and s_4 give rise to a transient which decays at the approximate rate $(\gamma_2 + \gamma_{21})/2$, and oscillates at approximately twice the effective two-photon Rabi frequency $2|V_{21}|$. This limit thus gives information on the dipole moments of the system. In Fig. 4.3c, we show the contributions to $\rho_{23}(t)$ due to s_3 and s_4 in the later limit. The spectrum exhibits two peaks at $\Delta_2 = \pm |V_{21}|$.



(a)



(b)



(c)

Fig. 4.3

Various contributions to the optical nutation signal at the Stark shifted two-photon resonance ($\Delta_1' = 0$). (a) Steady-state contribution, (b) transient contribution of the root s_2 and (c) of s_3 and s_4 when $|V_{21}| \gg |\gamma_2 - \gamma_{21}|/4$ (see Eq. 4.11). The relaxation rates are $\gamma_2 = 0.03$, $\gamma_3 = 0.77$, $\gamma_{21} = 0.1$, $\gamma_{23} = 0.5$, $\gamma_{13} = 0.49$, and the two-photon Rabi frequency $\Omega_{21} = V_{21} = 1$.

d. Similar comments apply to the roots s_5 and s_6 . Depending upon the relative magnitudes of $|V_{21}|$ and $|\gamma_{23} - \gamma_{13}|/4$, the square root in Eq. (4.11) yields either purely exponential decay or oscillations. In the

weak field regime, $|V_{21}| \ll |\gamma_{23} - \gamma_{13}|/4$, the s_5 and s_6 contributions to the transient signal decay at rates γ_{23} and γ_{13} , while oscillate at the beat frequency Δ_2' . For strong pump field, the transient signal decays at the average relaxation rate $(\gamma_{13} + \gamma_{23})/2$ and oscillates at the approximate superposition frequency $\Delta_2' \pm |V_{21}|$.

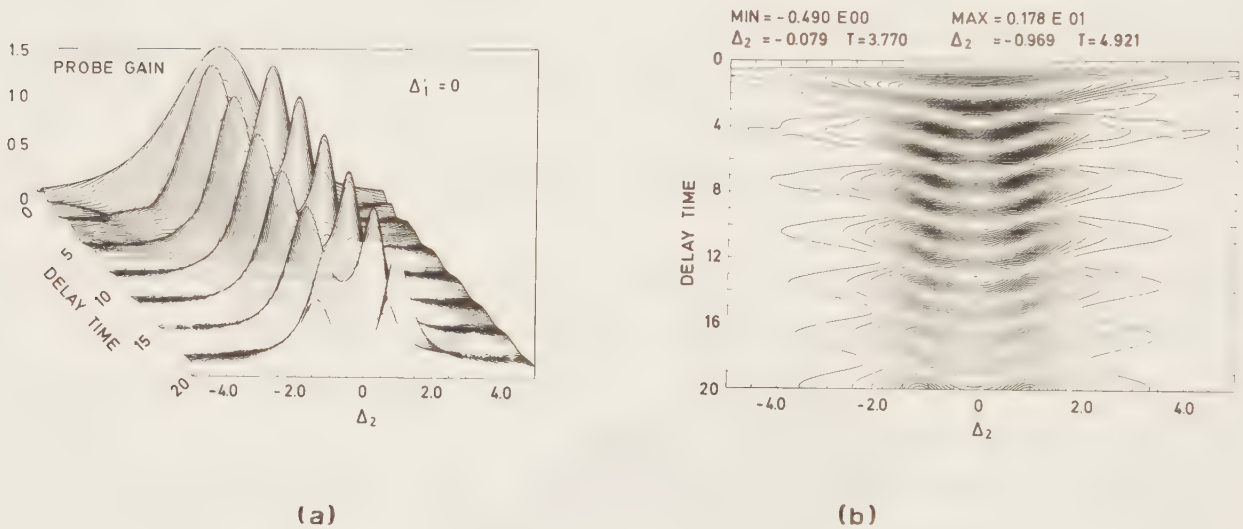


Fig. 4.4

(a) Transient probe gain at the Stark shifted resonance ($\Delta_1' = 0$) after the pump field is switched on, for the case $|V_{21}| = 1 > |\gamma_{23} - \gamma_{13}|/4$. Relaxation rates as in Fig. 4.3. (b) Corresponding contour plot.

In Fig. 4.4a, we show the total time dependence of the probe gain $\rho_{23}(t)$, in a case where the two-photon Rabi oscillation dominates over the atomic decays. We have chosen a case where the relaxation rate of level $|3\rangle$ is much faster than that of level $|2\rangle$ (consistently with our assumption $\gamma_3 \gg \gamma_2$). This is the case e.g. in Cadmium. Thus the contributions of s_2 , s_5 and s_6 decay away faster than those of s_3 and s_4 .

In Fig. 4.4b, we give the corresponding contour plot. It shows that for some range of time delays, the Stark splitting can be better

resolved than at steady state. Such features are well known in high-resolution spectroscopy and have been used in other contexts to achieve subnatural spectroscopy /62/.

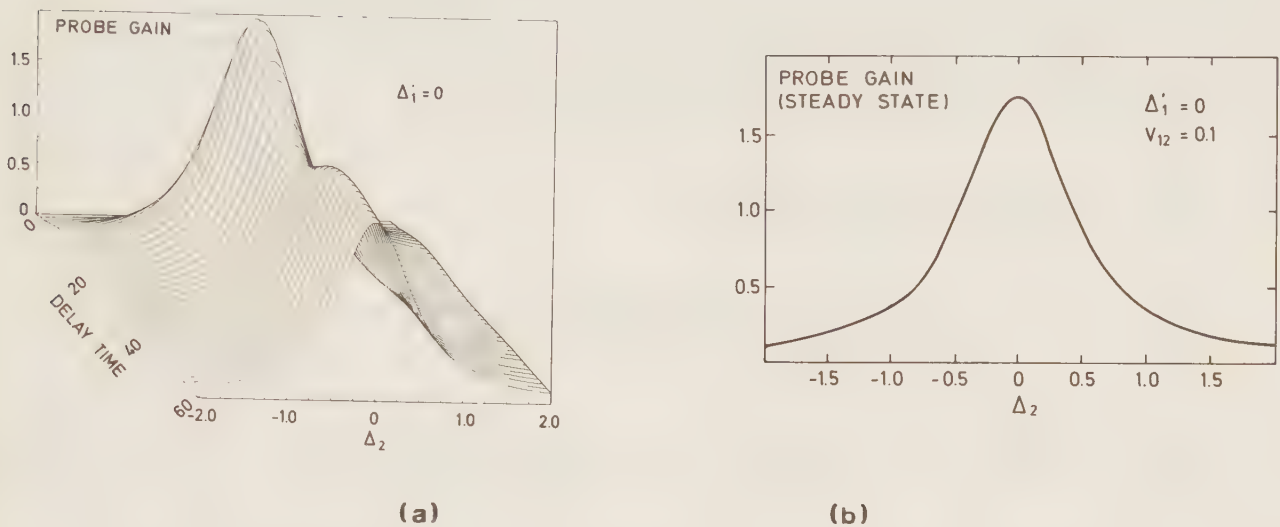


Fig. 4.5

(a) Optical nutation signal for weak pump fields. $|V_{21}| = 0.1 < \gamma_2 - \gamma_{21}/4$, $\Delta_1' = 0$. (b) Corresponding steady-state probe gain. Relaxation rates as in Fig. 4.3.

In Fig. 4.5a we contrast these results with the weak field limit, where essentially all oscillations disappear. (Oscillations introduced by s_5 and s_6 are short lived and of small magnitude.) We do not see any trace of Stark splitting (nor do we expect it); instead we observe the superposition of two signals: the first reaches its maximum approximately when the intermediate level transients have died out and decays at the two-photon coherence relaxation rate γ_{21} , while the second reaches its maximum at later times and decays at the rate γ_2 to the steady-state signal shown in Fig. 4.5b.

4.4.2. Perturbative approach in decay constants.

As a second example, we consider the limit of small decay constants, and specifically the case where the phase relaxation rate γ_{21} of the ρ_{21} coherence is much smaller than the Rabi frequency V_{21} . In the general case $\Delta_1' \neq 0$, the Laplace transforms of the matrix elements relevant for the determination of the probe gain $\rho_{23}(t)$ are

$$\begin{aligned}\bar{\rho}_{22}^{(0)}(s) &= \frac{2|V_{21}|^2 (s + \gamma_{21})(s + \gamma_3)}{s D_{21}(s)} \\ \bar{\rho}_{21}^{(0)}(s) &= - \frac{i V_{21} (s + \gamma_2) (s + \gamma_{21} - i \Delta_1') (s + \gamma_3)}{s D_{21}(s)}\end{aligned}\quad (4.13)$$

$$\bar{\rho}_{23}^{(1)}(s) = \frac{i V_{23}}{D_{23}(s)} \left\{ \bar{\rho}_{22}^{(0)} \left(1 - \frac{\gamma_2}{s + \gamma_3}\right) (s + \gamma_{13} - i \Delta_3') - i V_{21} \bar{\rho}_{12}^{(0)} \right\}$$

where the denominators $D_{21}(s)$ and $D_{23}(s)$ are the fourth and second order polynomials

$$D_{21}(s) = (s + \gamma_3) \left\{ (s + \gamma_2) \left\{ (s + \gamma_{21})^2 + \Delta_1'^2 \right\} + 2|V_{21}|^2 \left(2 + \frac{\gamma_2}{s + \gamma_3}\right) (s + \gamma_{21}) \right\}$$

and

$$D_{23}(s) = (s + \gamma_{23} + i \Delta_2') (s + \gamma_{13} - i \Delta_3') + |V_{21}|^2 \quad (4.14)$$

The Laplace transform $\bar{\rho}_{23}^{(1)}(s)$ has therefore seven poles. Approximated expressions for the poles associated with $D_{21}(s)$ may be obtained by rewriting D_{21} as

$$\begin{aligned}D_{21}(s) &= (s + \gamma_3) \left\{ (s + \gamma_2) \left\{ (s + \gamma_{21})^2 + \Delta_1'^2 + 4|V_{21}|^2 \right\} + 4|V_{21}|^2 \left(\gamma_{21} - \frac{\gamma_2}{2} \right) \right\} \\ &\quad + 2|V_{21}|^2 \gamma_2 (\gamma_{21} - \gamma_3)\end{aligned}\quad (4.15)$$

and assuming the last term to be small, so that it can be treated as

small perturbation. This approximation is again based in our assumption of a long lived upper state ($\gamma_2 \ll \gamma_3$). To zeroth-order in $2|V_{21}|^2 \gamma_2 (\gamma_{21} - \gamma_3)$, the first root is $s_1 = -\gamma_3$ and the remaining three roots can be obtained by standard methods. These solutions can then be used to find the first-order corrections by means of the Newton's expansion (4.10). To this order, the seven roots read (in order to simplify the results, we assume $\gamma_{21}, \gamma_2 \ll |V_{21}|$):

$$s_0 = 0$$

$$s_1^{(1)} = -\gamma_3 - \frac{c}{(-\gamma_3 + b + \frac{2q}{d}) \left((-\gamma_3 + b - \frac{q}{d})^2 + d \right)}$$

$$s_2^{(1)} = -b - \frac{2q}{d} - \frac{c}{(\gamma_3 - b - \frac{2q}{d}) \left((\frac{3q}{d})^2 + d \right)}$$

$$s_{3/4}^{(1)} = -b + \frac{q}{d} \pm i\sqrt{d} - \frac{c}{(\gamma_3 - b + \frac{q}{d} \pm i\sqrt{d}) \left((\frac{3q}{d} \pm i\sqrt{d}) (\pm 2i\sqrt{d}) \right)}$$

$$s_5 = -\frac{\gamma_{13} + \gamma_{23}}{2} - i\frac{\Delta_2 - \Delta_3}{2} \pm \frac{i}{2} \sqrt{4|V_{21}|^2 - (\gamma_{23} - \gamma_{13} + i\Delta_1)^2} \quad (4.16a)$$

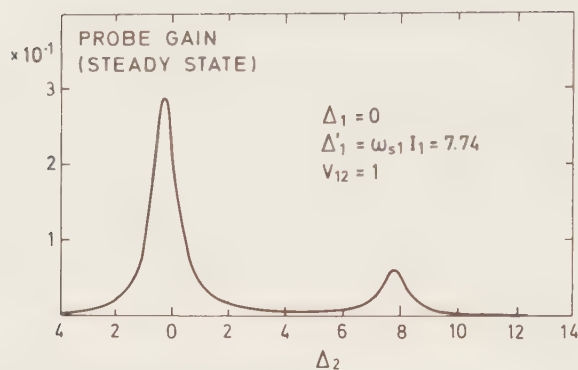
where

$$b = \frac{\gamma_2 + 2\gamma_{21}}{3}$$

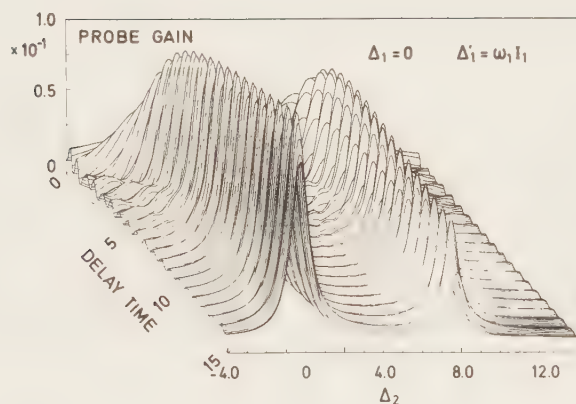
$$q = \frac{\gamma_2 - \gamma_{21}}{3} \left[\Delta_1'^2 + \left(\frac{\gamma_2 - \gamma_{21}}{3} \right)^2 \right] + |V_{21}|^2 \frac{\gamma_2 + \gamma_{21}}{3}$$

$$d = 4|V_{21}|^2 + \Delta_1'^2 - \frac{(\gamma_2 - \gamma_{21})^2}{3}$$

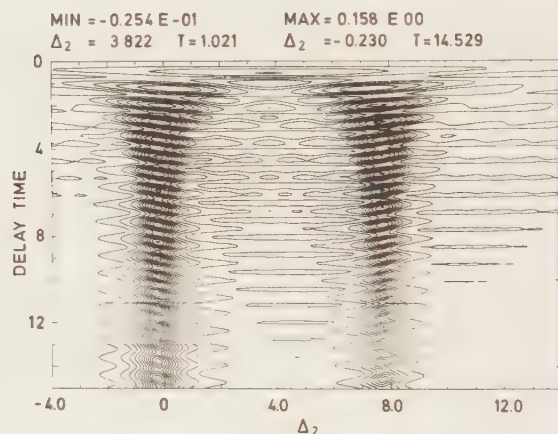
$$c = 2|V_{21}|^2 \gamma_2 (\gamma_{21} - \gamma_3). \quad (4.16b)$$



(a)



(b)



(c)

Fig. 4.6

Transient probe gain at the two-photon resonance ($\Delta_1=0$). (a) Steady-state contribution, (b) time dependent gain spectrum and (c) its corresponding contour plot. Relaxation rates as in Fig. 4.3.

We checked these results by comparing the product of the factorized polynomial with the exact one {Eq. (4.15)}. Fig. 4.6a shows the steady-state probe gain for $\Delta_1=0$, $\Delta_1'=\omega_{s1} I_1=7.74 \text{ nsec}^{-1}$. As expected, it consists of two lines: one at the two-photon resonance, and the Raman line located at $\Delta_2=\Delta_1'$. The corresponding time dependent gain spectrum

$\rho_{23}^{(1)}(t, \Delta_2)$ is plotted in Fig. 4.6b, and its contour plot is shown in Fig. 4.6c. The short time behavior is characterized by a broad Lorentzian centered about half-way between these two lines (see the contour plot). For longer times, both lines become resolved and eventually reach the steady-state limit. Our results are only valid for relaxation rates smaller than the effective two-photon Rabi frequency. Therefore we are always in the regime where $s_3^{(1)}$, $s_4^{(1)}$, s_5 and s_6 give oscillatory contributions. Use has been made of our assumption $\gamma_3 \gg \gamma_2$ so that the transient introduced by $s_1^{(1)}$ decays faster than those introduced by $s_2^{(1)}$, $s_3^{(1)}$ and $s_4^{(1)}$. The plots correspond to the case $\gamma_{21} < \gamma_{23}, \gamma_{13}$, in which the transient signals originating in s_5 and s_6 die out faster than those decaying with the ρ_{21} coherence relaxation rate. For times about the inverse of the average relaxation rate $(\gamma_{13} + \gamma_{23})/2$, the intermediate state transients die out, and both the population- and the Raman-type line become resolved. From there on, the population line increases while the Raman line decreases to their steady-state values. This part of the transient regime is determined by the roots s_2 and $s_{3,4}$. The time oscillations are due only to $s_{3,4}$ and have the frequency $\sqrt{4|V_{21}|^2 + \Delta_1'^2}$.

For this example, where $\Delta_1 = 0$, the effective detuning Δ_1' is due only to the Stark shift. The Raman line is centered at $\Delta_2 = \omega_{S1} I_1$, and the oscillations occur to the frequency $\sqrt{4|V_{21}|^2 + (\omega_{S1} I_1)^2}$. Thus the probe gain allows to determine both the effective two-photon Rabi frequency and the Stark shift of the system under study. The observation of the optical nutation may be also an efficient means to determine experimentally the Stark shifted resonance. The minimum oscillation frequency of the transient signal occurs at the Stark shifted resonance, and coincides approximately with twice the two-photon effective Rabi frequency.

4.5 Free induction decay

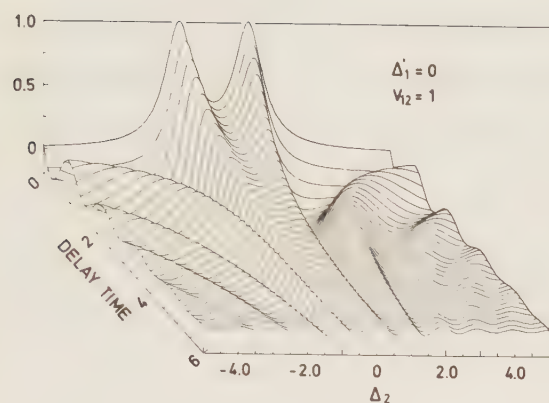
The expression 'free induction decay' (FID) is used here rather loosely, in the sense that while the strong pump field is switched off at $t=t_0$, as in usual FID experiments, the probe field remains applied to the system. The free pump field solution is obtained from Eqs. (4.5) and (4.6) by setting V_{12} to zero. The probe gain spectrum is then given by the imaginary part of

$$\begin{aligned} \rho_{23}(t) = & \rho_{23}(t_0) e^{-(\gamma_{23} + i\Delta_2)(t-t_0)} \\ & + iV_{23} \left\{ \rho_{22}(t_0) \frac{\gamma_3 - 2\gamma_2}{\gamma_3 - \gamma_2} \frac{e^{-\gamma_2(t-t_0)} - e^{-(\gamma_{23} + i\Delta_2)(t-t_0)}}{\gamma_{23} + i\Delta_2 - \gamma_2} \right. \\ & \left. - \left(\rho_{33}(t_0) - \frac{\gamma_2}{\gamma_3 - \gamma_2} \rho_{22}(t_0) \right) \frac{e^{-\gamma_3(t-t_0)} - e^{-(\gamma_{23} + i\Delta_2)(t-t_0)}}{\gamma_{23} + i\Delta_2 - \gamma_3} \right\} \end{aligned} \quad (4.17)$$

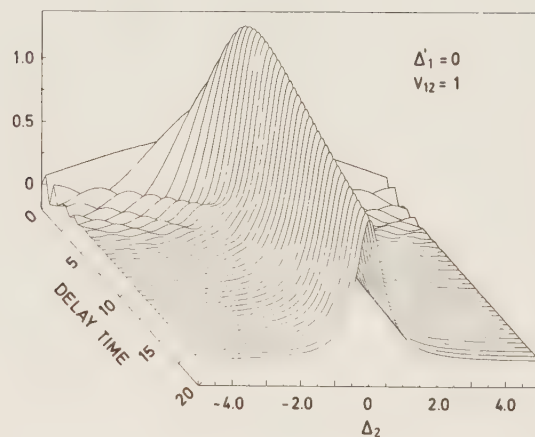
This expression contains three contributions: the standard free decay of the coherence $\rho_{23}(t_0)$ at the rate γ_{23} ; and the couplings of the initial populations $\rho_{22}(t_0)$ and $\rho_{33}(t_0)$ with the probe field. Fig. 4.7a shows the free decay of $\rho_{23}(t_0)$ at the Stark shifted two-photon resonance ($\Delta_1' = 0$). It is assumed for simplicity that the pump field is switched off after the system has reached steady-state, i.e., $\rho_{23}(t_0)$, $\rho_{22}(t_0)$ and $\rho_{33}(t_0)$ are the steady-state values studied in Chapter III. We can see how the Stark splitting dies out with oscillations due to the detuning Δ_2 .

The contribution of the population term $\rho_{22}(t_0)$ (second term in Eq. (4.17)) is illustrated in Fig. 4.7b. At $t=t_0$ it is zero, then starts growing around $\Delta_2 = 0$ up to a maximum at the delayed time

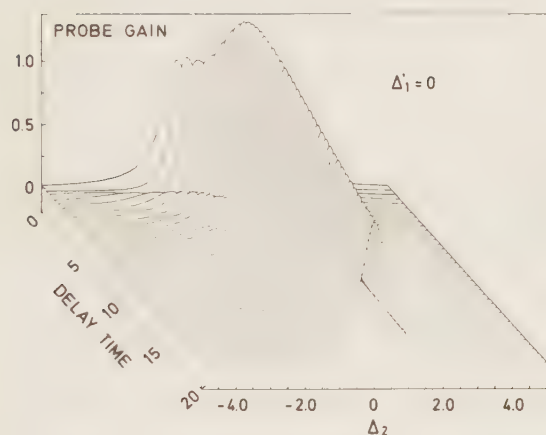
$$T = t - t_0 = \frac{\ln \delta_{23} - \ln \delta_2}{\delta_{23} - \delta_2}. \quad (4.18)$$



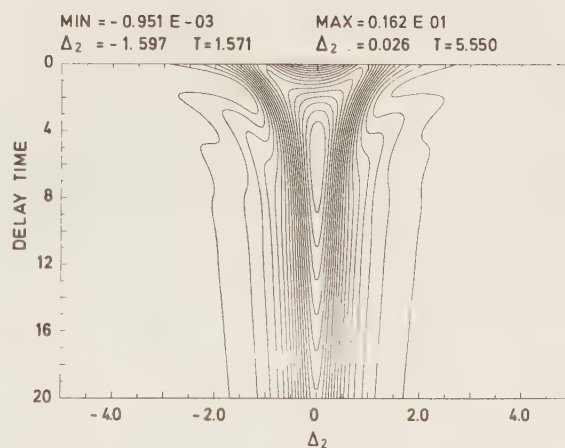
(a)



(b)



(c)



(d)

Fig 4.7

Free induction decay at the Stark shifted resonance ($\Delta_1' = 0$). $V_{21} = 1$ ($t < t_0$) and relaxation rates as in Fig. 4.3. (a) Free decay of the coherence $\delta_{23}(t_0)$ (first term in Eq. (4.17)). (b) Contribution of the population term $\delta_{22}(t_0)$ (2nd. term in Eq. (4.17)). (c) Total free induction decay signal and (d) its corresponding contour plot.

and then decreases at the population decay rate γ_2 . The contribution of the third term is of the order of γ_2/γ_3 times that of the second and therefore negligible compared to it. The total gain spectrum is plotted in Fig. 4.7c. Because of $\gamma_3 \gg \gamma_2$, $\gamma_{23} \geq (\gamma_2 + \gamma_3)/2 \gg \gamma_2$ so that the long time transient is determined by the $\rho_{22}(t_0)$ contribution, whereas for short delay times the free decay of $\rho_{23}(t_0)$ predominates.

The corresponding contour plot (Fig. 4.7d) further illustrates the relaxation of the Stark splitted spectrum to a Lorentzian. The maximum of the total gain spectrum is approximately reached at the delay time given by Eq. (4.18).

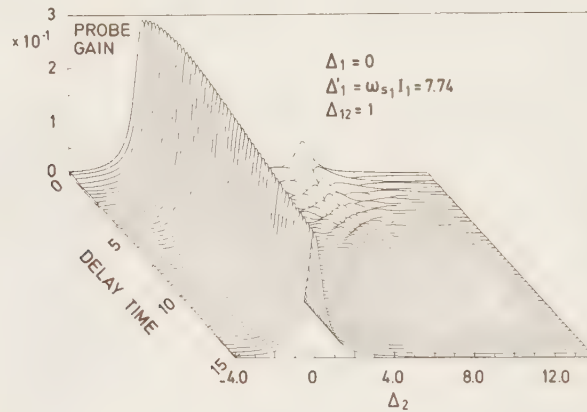


Fig. 4.8

Gain spectrum at the two-photon resonance ($\Delta_1 = 0$).
Relaxation rates as in Fig. 4.3.

Fig. 4.8 shows the gain spectrum when pumping at $\Delta_1 = 0$, i.e., $\Delta'_1 = \omega_{s1} I_1$. After reaching a maximum, the population signal decays with γ_2 , while the Raman line, centered at $\Delta_2 = \Delta'_1$, relaxes like the coherence with the relaxation rate γ_{23} .

4.6 Conclusions

We have divided our analysis into two cases: resonant and nonresonant pumping. The Stark shift acts as a pump intensity dependent detuning; therefore, to obtain the spectra corresponding to the resonance, one should pump at $\Delta_1 = -\omega_{S1} I_1$, so that $\Delta_1' = 0$. If pumping is performed at the real resonance $\Delta_1 = 0$, the spectrum corresponds to $\Delta_1' = \omega_{S1} I_1$, and its shape corresponds to the second case.

We have considered the case $\gamma_2 \ll \gamma_3$, which for small pressure broadening implies $\gamma_2, \gamma_{21} < \gamma_3, \gamma_{23}, \gamma_{13}$, so that the transient signals related with the intermediate states die out much faster than those related with the two-photon coherence ρ_{21} or the population ρ_{22} in the upper state.

At the Stark shifted resonance, we have studied the optical nutation signal in the weak and strong field regime. For weak fields ($|V_{21}| \ll |\gamma_2 - \gamma_{21}|/4$), we observed a rapid rise of a signal centred at $\Delta_2 = 0$ followed by exponential decay at rates γ_2 and γ_{21} .

The study of the transient gain spectrum for strong fields ($|V_{21}| \gg |\gamma_2 - \gamma_{21}|/4$) illustrated how the Stark splitting first builds up in times comparable to the inverse of the intermediate state relaxation rates. Thereafter the gain maxima approach steady-state showing damped oscillations at the frequency $2|V_{21}|$ and damping constant $(\gamma_{21} + \gamma_2)/2$.

The transient gain spectrum for free induction decay after steady-state preparation enabled us to see how the Stark splitting induced by the strong pump field dies out at the rate γ_{23} , while a delayed strong gain signal at $\Delta_2 = 0$ -originating in the coupling between the upper state population and the weak probe field- builds up, reaches a maximum at a delayed time approximately given by Eq. (4.18), and then

relaxes at the upper level population relaxation rate γ_2 . The maximum value of the signal depends on the value of the steady-state population in the upper level.

For nonresonant pumping, we found approximate, perturbative solutions in relaxation rates, i.e., for fields strong enough that $\gamma_2, \gamma_{21} \ll |V_{21}|$. For pump detunings larger than the effective two-photon Rabi frequency, we can distinguish between population-type and Raman-type signals. As the pump is switched-on, both signals first increase, but after the transients related to the intermediate state die out, the population-type signal still increases, whereas the Raman line decreases, both towards their steady-state value. Both lines oscillate at the frequency $\sqrt{4|V_{21}|^2 + \Delta_1^2}$. If the pump field is switched-off after the system has reached steady-state, the Raman line relaxes at the relaxation rate of the coherence ρ_{23} . The inclusion of the probe gives rise to the long time transient signal of the population, and a possibility to measure the population decay rate γ_2 . Otherwise one would only observe the free decay of $\rho_{23}(t_0)$ at the rate γ_{23} .

In Figs. 4.3 to 4.8, we also included the Stark shift $\omega_{32}I_1$. Its effect is however very small here because its value is very small (we have used the values for Cd). If the $\omega_{32}I_1$ shift were larger, its effect would be just to shift the total optical nutation gain spectrum, while its relaxation would be seen in the free induction decay spectrum. Nevertheless, it is to be expected that shifts of the upper and intermediate level be of the same order so that the relative shift between states $|2\rangle$ and $|3\rangle$ remains small.

CHAPTER V

GAUSSIAN PUMP PULSES AND PULSE PROPAGATION

5.1 Introduction

Having studied in the preceding chapters the gain of a weak probe field both at steady-state and for a square pump pulse, we now turn to the more realistic case of a Gaussian pump pulse. The fact that the pump intensity varies now in a finite time from zero to a maximum and back opens new questions: (i) How does the transient probe gain for Gaussian pulses behave? (ii) How does the Stark shift affect its temporal shape? (iii) Under which conditions is it possible to describe the system as if it were at steady-state?

From our study of transients in Chapter IV, we know that the strongest Raman signal is obtained when the pump field is switched on (see Fig. 4.6b); it decreases thereafter to its steady-state value (Fig. 4.6a). On the other hand, there is an increase in the population signal after the pulse is switched off (see Fig. 4.8). During the pulse, when working at the Stark shifted resonance where the gain is largest, the steady-state regime is reached for times long compared to the inverse of $(\gamma_2 + \gamma_{21})/2$.

In the first part of this chapter, we present temporal gain shapes for Gaussian pulses of nanosecond duration and of high enough intensity for the effective two-photon Rabi frequency to be of the order of nsec^{-1} . Working first within the thin sample approximation, we give examples for slow decay rates (on the order of tenths of nsec^{-1}), and then for faster ones (ten nsec^{-1}). We consider in this way two different

regimes: in the first the pump field is strong, i.e. its Rabi frequency is larger than the relaxation rates, and rapid transients dominate. In the second the opposite situation prevails and polarizations are expected to follow adiabatically the field variations; transient effects become unimportant, except for the decay of the population ρ_{22} at rate γ_2 after the pump field is switched off.

Another interesting problem to be analyzed here is the effect of the Stark shifts. In a Gaussian pulse, the pump intensity is not constant, so that Stark shifts lead to a time-dependent detuning. The upper level $|2\rangle$ is first pushed away from resonance during the leading edge of the pump pulse, to then return gradually to its unperturbed position. This can introduce additional characteristics in the temporal shape of the hyper-Raman signal.

We devote the second part of this chapter to the study of the propagation of Gaussian pulses through an extended medium. Here, we release the thin sample approximation and solve numerically the coupled field and atomic equations of motion. For simplicity we formulate the SVEA in a slightly different way as in Chapter II; we then present numerical results for special cases, and finally compare qualitatively our predictions with the experimental results of ref. /12/.

5.2 Probe gain for a Gaussian pump pulse

In this section, we assume as in Chapter IV that a weak field probes the transition $|2\rangle \leftrightarrow |3\rangle$ (see Fig. 4.1a). We look for the temporal behavior of its gain as a function of time and probe or pump detuning. We first make the thin sample approximation (see Section 4.2), so that the gain is simply proportional to the imaginary part of $\rho_{23}(t)$.

evaluated in terms of the input fields. We solve the density matrix equations numerically with an Adams-Bashforth code (see e.g. /61/).

5.2.1. Slow decay rates.

Consider first the situation where all decay rates are small compared to the maximum effective two-photon Rabi frequency ($|V_{21}^{\max}|$). Using the same decay constants as in the preceding chapter, which correspond to a moderate homogeneous broadening in Cd, we solve the density matrix equations for a Gaussian pump pulse of duration on the order of the inverse of the phase relaxation rates. The probe field is taken to be constant, and with a Rabi frequency V_{23} much smaller than the maximum two-photon effective Rabi frequency.

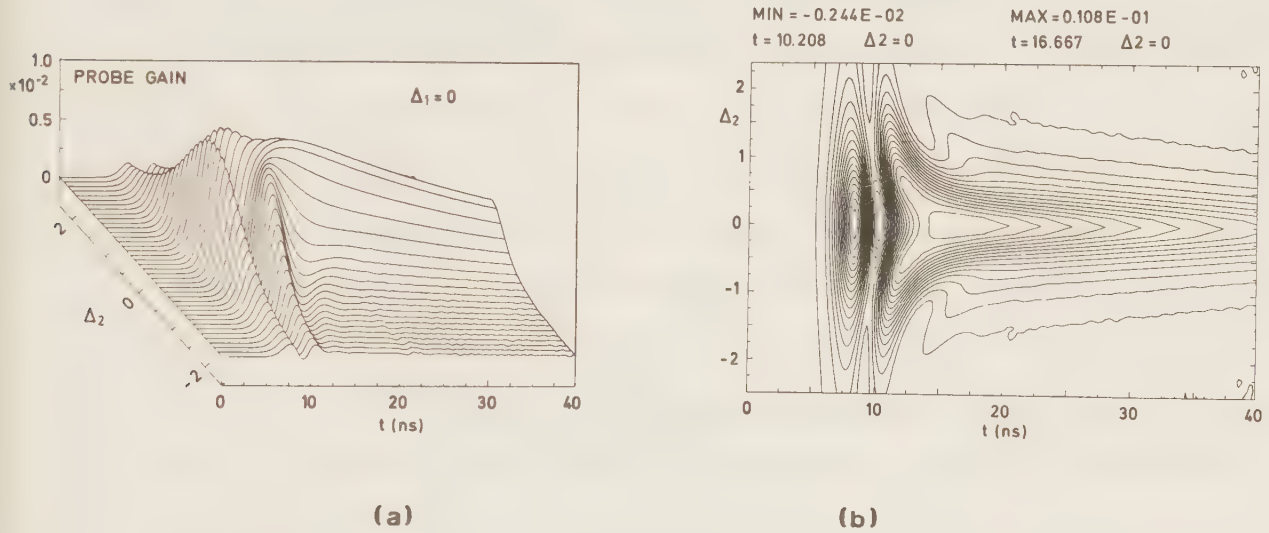


Fig. 5.1

(a) Probe gain spectrum at the two-photon resonance ($\Delta_1 = 0$) as a function of time and probe detuning. The Stark shifts have been arbitrarily set to zero. (b) Corresponding contour plot. The relaxation constants are $\gamma_2 = 0.03 \text{ ns}^{-1}$, $\gamma_3 = 0.77 \text{ ns}^{-1}$, $\gamma_{21} = 0.1 \text{ ns}^{-1}$, $\gamma_{23} = 0.5 \text{ ns}^{-1}$, $\gamma_{13} = 0.49 \text{ ns}^{-1}$; $V_{21}^{\max} = 1 \text{ ns}^{-1}$. The pump pulse is a gaussian of 6-ns width centered at $t = 9$ ns.

If we set the Stark shifts arbitrarily to zero, and probe the two-photon resonance ($\Delta_1=0$) by tuning the probe, we obtain the temporal shapes shown in Fig. 5.1a. The maximum gain is reached at resonance ($\Delta_2=0$) and after the pulse is over ($t=16.67$ nsec). This is clearly shown in the corresponding contour plot (Fig. 5.1b). This maximum is just the population transient of free induction decay found in Chapter IV, and has a long tail decaying at the population decay rate γ_2 . During the pump pulse, the gain spectrum is reminiscent of the optical nutation signal. We observe Rabi oscillations as well as the build-up of the Stark splitting.

In Fig. 5.2a we show the temporal behavior of the probe gain when the Stark shifts are included and the pump detuning Δ_1 varied. The probe field is tuned at resonance ($\Delta_2=0$). For $\Delta_1=-\omega_{s1}I_1^{\max}=-7.74$ nsec $^{-1}$, where $I_1^{\max}=|V_{21}^{\max}|2\sqrt{T_1/\gamma_{21}}$, as defined in Chapter II, the population line is much weaker than that at $\Delta_2=0$ in Fig. 5.1a. It means that one cannot compensate the effect of the Stark shift by detuned pumping at $\Delta_1=-\omega_{s1}I_1^{\max}$, as in the case of square pump pulses. Comparing Fig. 5.1 and 5.2 shows that an approximate compensation of the dynamic Stark shift is obtained for $\Delta_1=-5.47$. This corresponds to the Stark shift value $\Delta_1=-(1/\sqrt{2})\omega_{s1}I_1^{\max}$, rather than to the average value $\Delta_1^{\text{av}}=-(\sqrt{\pi}/2)\omega_{s1}I_1^{\max}$ for a Gaussian pulse (*). This value of Δ_1 gives a rough idea of the 'effective Stark shift', which must be compensated by a real detuning to reproduce the resonance characteristics obtained in the absence of the Stark shifts.

$$(*) I_1^{\text{av}} = 1/2\gamma \int_{-\infty}^{\infty} dt I_1^{\max} \exp(-t/\gamma)^2 = (\sqrt{\pi}/2) I_1^{\max}.$$

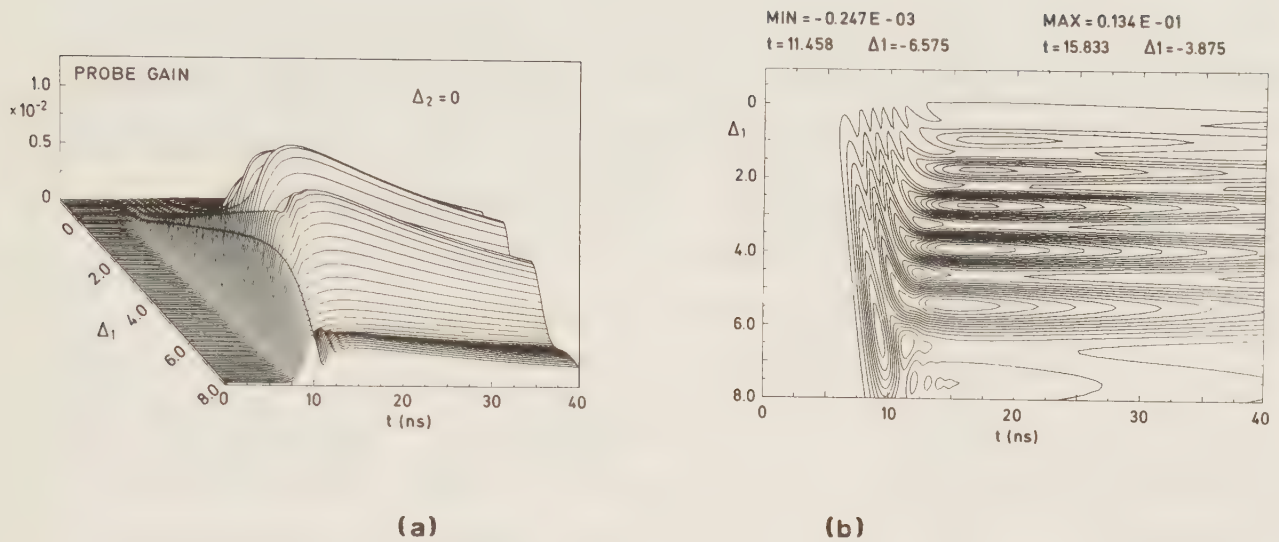


Fig. 5.2

(a) Gain at $\Delta_2 = 0$ as function of time and pump detuning when Stark shifts are included. (b) Corresponding contour plot. Relaxation rates and pulse shape as in Fig. 5.1.

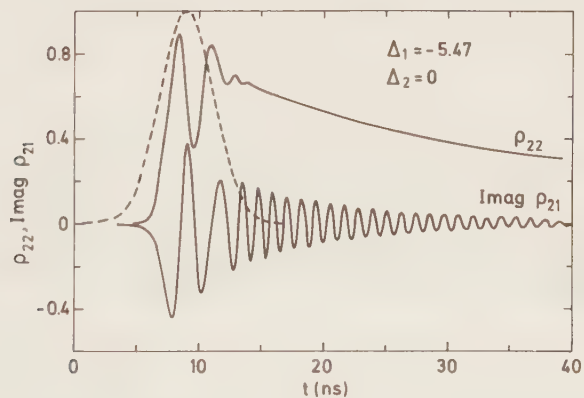


Fig. 5.3

Upper state population ρ_{22} and imaginary part of the two-photon coherence ρ_{21} as function of time for $\Delta_1 = -5.47$ and $\Delta_2 = 0$. The pump pulse is shown in dotted line. Relaxation rates as in Fig. 5.1.

In Fig. 5.3 we show the upper state population ρ_{22} and the imaginary

part of the two-photon coherence ρ_{21} at $\Delta_1 = -5.47$ (which, as we have seen, corresponds to a rough compensation of the dynamic Stark shift) and $\Delta_2 = 0$. During the pump pulse ρ_{22} and ρ_{21} oscillate at approximately twice the maximum Rabi frequency. These Rabi oscillations are typical of optical nutation. After the pulse is over, the population decays at the rate γ_2 , while the two-photon coherence ρ_{21} undergoes decaying oscillations at the frequency of the real detuning Δ_1 .

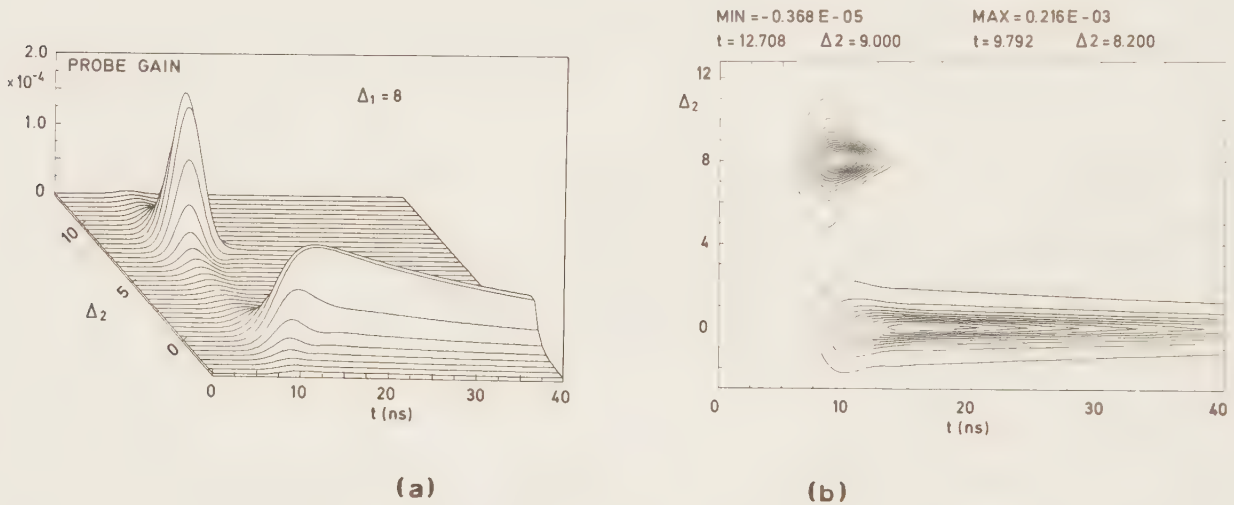


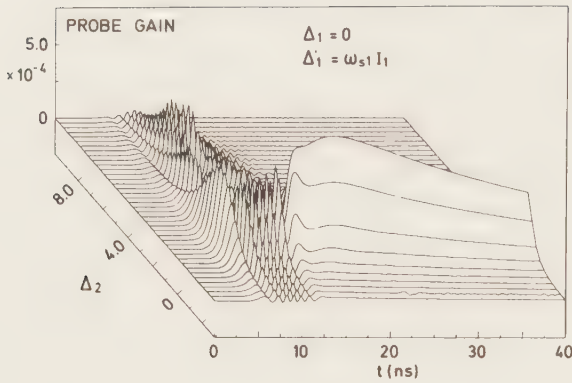
Fig. 5.4

(a) Probe gain spectrum as function of time for large pump detuning $\Delta_1 = 8$, when the Stark shifts are arbitrarily set to zero. (b) Corresponding contour plot. Pump pulse and relaxation rates as in Fig. 5.1. The distinction between Raman and population signal is evident.

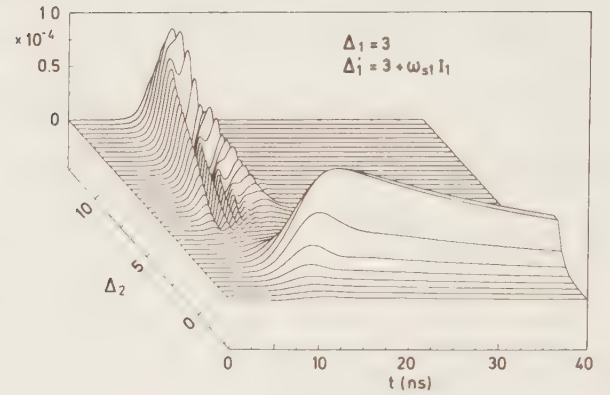
Let us now discuss the behavior of the probe gain as a function of time and of the probe detuning Δ_2 . For large pump detunings, we expect two lines to appear in the probe gain spectrum: a population line centered at $\Delta_2 = 0$, and a Raman line centered about $\Delta_2 = \Delta_1$. Fig. 5.4a shows the temporal behavior of these two lines for $\Delta_1 = 8$ and the Stark shifts arbitrarily set to zero. As the contour plot (Fig. 5.4b) shows, the two signals are clearly separated in the time-frequency plane. The Raman

line, centered around $\Delta_2 = \Delta_1 = 8$, is about 1.5 times larger than the population signal and, as expected, it exists only during the pump pulse. The population line reaches its maximum after the pump pulse is over at around $t = 16$ nsec. This behavior is related to the free induction decay.

When Stark shifts are included the gain spectrum becomes more complicated. In Fig. 5.5a we show the gain spectrum when pumping at the unshifted two-photon resonance $\Delta_1 = 0$, so that $\Delta_1' = \omega_{s1} I_1$. Instead of the two clearly separated lines, a Raman line around $\Delta_2 = \Delta_1'$ and a population line around $\Delta_2 = 0$, we see a strongly oscillating signal during the laser pulse, and a population signal centered around $\Delta_2 = 0$ with its maximum appearing after the pump pulse. The population signal resembles that of Fig. 5.4a. This is to be expected, since the population contribution depends mainly on the population of the upper state at the end of the pulse. The behavior during the pulse can be understood intuitively as follows: at the beginning of the pulse, the intensity is too small to shift considerably the levels. The gain spectrum has then only one maximum near $\Delta_2 = 0$. As time goes on, the pump intensity increases, the levels are pushed away and an oscillating Raman signal shows up around $\Delta_2 = \Delta_1'$. Contrary to the case of Fig. 5.4a this signal is smaller than the population signal at the end of the pulse. This is not surprising: by setting the Stark shifts to zero, we keep the atomic levels fixed, so that the pump detuning is constant. In real fact, however, the pump pulse pushes level $|2\rangle$ away and back producing a time-dependent detuning, which varies in time from zero to $\Delta_1' = \omega_{s1} I_1^{\max}$.



(a)



(b)

Fig. 5.5

Probe gain spectrum when Stark shifts are included.
 (a) At $\Delta_1 = 0$, so that $\Delta_1' = \omega_{s1} I_1$, and (b) at $\Delta_1 = 3$.
 Pump pulse and relaxation rates as in Fig. 5.1.

Fig. 5.5b shows the gain spectrum as a function of time when a finite pump detuning is present in addition to the Stark shift. Its structure is already closer to that of Fig. 5.4 with a Raman line with maximum around $\Delta_2 = \Delta_1'$ which can be clearly distinguished from the population line at $\Delta_2 = 0$. This is because in this case, the maximum Stark shift is comparable to the fixed detuning Δ_1 . Indeed, as would be expected, as the fixed detuning is further increased, the role of the time-dependent Stark detuning becomes less important, and can be neglected for $\Delta_1 \gg \omega_{s1} I_1$.

5.2.2. Fast decay rates.

As already mentioned at the beginning of this chapter, when the decay rates are much larger than the maximum Rabi frequency and the inverse of the pulse duration, the transients are effectively damped out. In Fig. 5.6 we show the Stokes gain for transversal decay rates much larger than

the two-photon effective Rabi frequency, and in Fig. 5.7 the corresponding upper-state population ρ_{22} and the two-photon coherence ρ_{21} (multiplied by a factor 10).

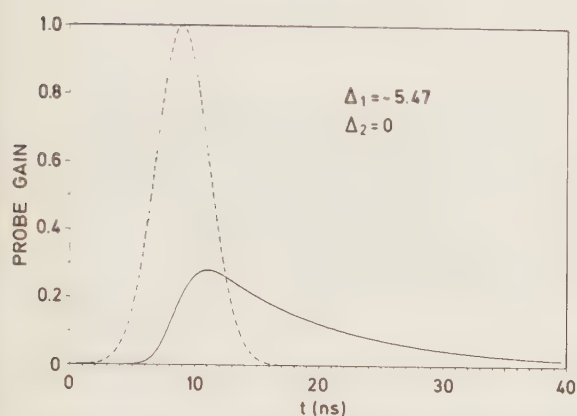


Fig. 5.6

Temporal shape of the probe gain for fast phase relaxation rates. Stark shifts have been included. The relaxation rates are $\Gamma_2 = 0.1$, $\gamma_3 = 1$, $\gamma_{21} = \gamma_{23} = \gamma_{13} = 10$. The pump pulse (in dotted line) is the same as used in Fig. 5.1.

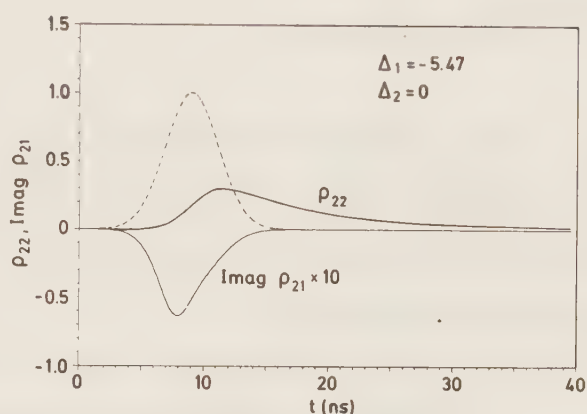


Fig. 5.7

Upper state population and imaginary part of the two-photon coherence ρ_{21} , as functions of time for the parameters of Fig. 5.6.

Neither the Stokes gain nor the population exhibit oscillations. The upper-state population reaches roughly the steady-state value it would reach for a square pump pulse of intensity equal to $(1/\sqrt{2})$ the maximum of the Gaussian pulse. Its maximum occurs after the pump pulse maximum, while the minimum of ρ_{21} occurs before. The fact that the system has now time to reach steady-state values during the pump pulse explains why a good qualitative agreement with the experimental measurements can be obtained from a steady-state Stokes gain calculation. Even the gain growth after the pulse due to ρ_{22} becomes less important here, because the delay time needed to reach its maximum is now very short (see Eq. (4.18)). The gain maximum turns out to be very close to the

steady-state value. Only the long tail decaying at a rate γ_2 reminds us of the transient character of the population signal.

5.3 Pulse propagation

In this section, we relax the thin sample approximation and consider the propagation of the Stokes and idler fields through an extended medium. This enables us to study the build-up of the fields for an arbitrary pump pulse.

Pulse propagation problems can be handled numerically rather efficiently by working in a frame moving at the speed of the pulse -the so-called retarded frame- where the variables z and t are replaced by $\xi=z$ and $\tau=t-z/c$. In order to take full advantage of this method we need to modify the formalism of Chapter II slightly. Specifically, we now expand fields and polarizations as plane waves, but using the vacuum wave-vectors instead of the medium wave-vectors (cf. Eq. (2.1) and (2.16)). As a consequence the phase mismatch Δk in Eq. (2.11) disappears. Furthermore, we now perform the SVEA by assuming that the envelope of the total polarization, and not just that of the nonlinear one, is slowly varying in time (see Eq. (2.21)). Finally, we neglect for simplicity the background polarization. In this way, we recover Eq. (2.27) for the field envelopes, but with the light velocity in vacuum rather than in the medium. This enables us to define an unique retarded time τ . The transformation from the independent variables (z,t) to (ξ,τ) is accomplished then by

$$\xi = z$$

$$\tau = t - z/c$$

$$\partial/\partial\tau = \partial/\partial t$$

$$\partial/\partial\xi = \partial/\partial z + 1/c \partial/\partial\tau \quad (5.1)$$

The equations we have then to solve are the matter equations (2.11) (where the left hand side derivative is with respect to the retarded time τ), and the field equations (2.27) where the derivative is with respect to ξ . These matter-field equations are solved simultaneously by numerical integration. For this purpose, we further developed a computer code used in /14/ by replacing the two-point extrapolation by a four-point predictor-corrector method.

The initial values of the fields are given at $z=\xi=0$, while the density matrix elements ρ_{ij} are initialized along the characteristics $\tau=0$. One refers to this way to implement initial conditions as the swept-excitation amplifier conditions, because a given slice of active medium does not start evolving until the various fields reach it, and it is therefore implicitly assumed that the medium at z is prepared in its initial state at $t=z/c$. In practice the swept-gain conditions are an excellent approximation if the life-time of the initial state is long compared to the propagation time of the pulses through the medium, and is therefore exact for atoms at rest initially in the ground state.

For conciseness, we limit our presentation of numerical results to a single, particular case representing a summary of the results of Chapter III and IV for the case of large phase relaxation rates. (In order to keep the computer time within reasonable limits, it was necessary to consider atomic densities several orders of magnitude lower than those

typically used in experiments). The sequence of Figs. 5.8a to 5.8d shows the various fields at several points along the amplifier, while Figs. 5.9a to 5.9d show the corresponding populations ρ_{ij} . For ease of discussion, we have also plotted the shape of the incident pump field (dotted lines).

Fig. 5.9a shows the populations near the beginning of the amplifier as a function of the retarded time τ . At the beginning of the pulse, the system is in the ground state and evolves to the steady-state population values, which are practically reached during the trailing edge of the pulse. The values we have used for the population relaxation rates correspond to those of Cd, and satisfy $\gamma_2 \ll \gamma_3$, which means that level $|2\rangle$ decays slowly to $|3\rangle$ which then transfers almost instantly its population into the ground state $|1\rangle$. This can be observed in Fig. 5.9a after the pump pulse: the upper state decays at its relaxation rate into $|3\rangle$ which transfers it at the rate γ_3 , more than 20 times larger than γ_2 , into the ground state. The dynamics of the density matrix elements is essentially determined by the pump pulse alone.

At the beginning of the amplifier, the Stokes and idler fields grow exponentially, i.e., in the small gain regime. Their early Rabi frequencies are shown in Fig. 5.8a. The Stokes Rabi frequency exhibits a maximum during the trailing edge of the pump pulse and some small oscillations (not observable in the Figure) during the leading edge of the pump pulse. Interesting is the fact that the idler field has two maxima, a large one at the beginning of the pump pulse, and a weaker, broader one during its second half. The first maximum is clearly related with an optical nutation transient, while the weaker is a population transient.

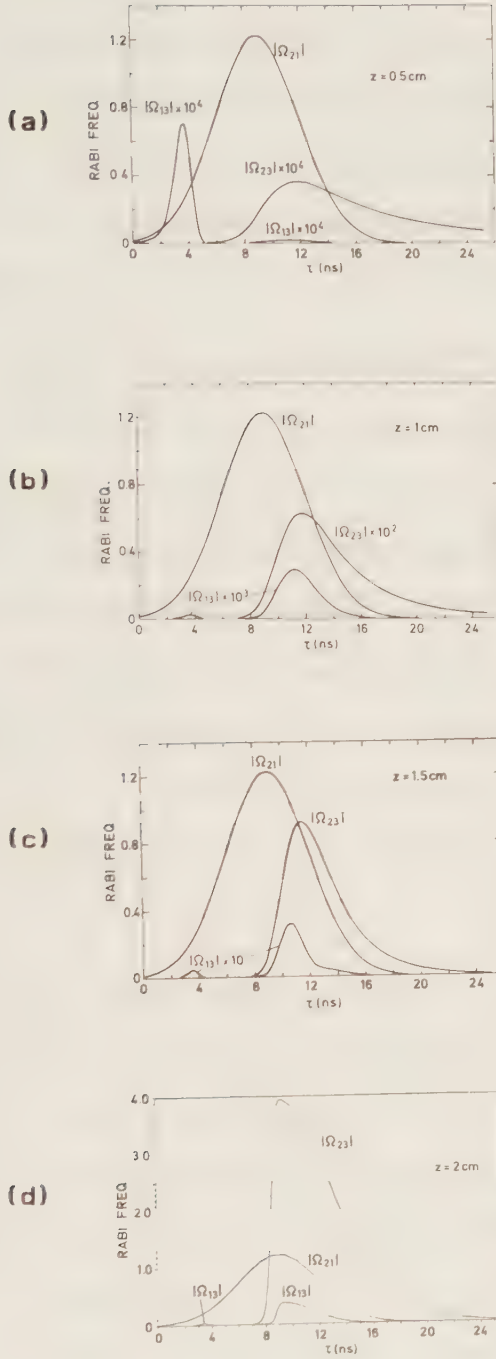


Fig. 5.8

Pump (Ω_{21}), Stokes (Ω_{23}) and idler (Ω_{13}) Rabi frequencies as functions of time along the amplifier at (a) $z=0.5 \text{ cm}$, (b) $z=1 \text{ cm}$, (c) $z=1.5 \text{ cm}$, (d) $z=2 \text{ cm}$. Relaxation rates $\gamma_2 = 0.03$, $\gamma_3 = 0.77$, $\gamma_{12} = \gamma_{23} = \gamma_{13} = 10 \text{ ns}^{-1}$. $\Delta_1 = -0.7 \omega_{21} I_{\text{max}}$. Stark shifts and dipole moments as for Cd. (see Chapter III).

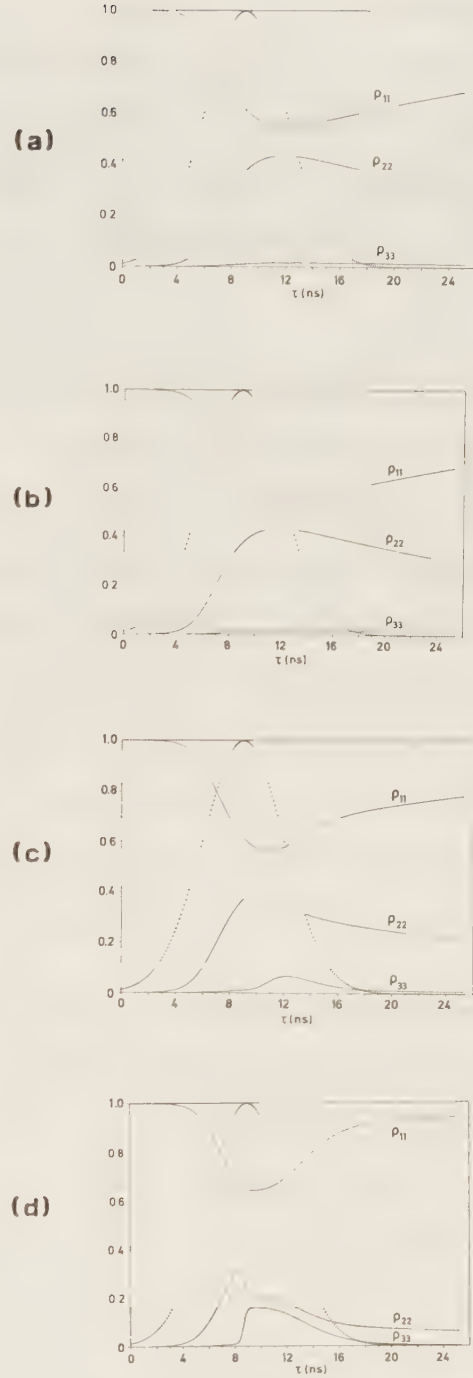


Fig. 5.9

Population and pump pulse (dotted line) along the amplifier. (a) $z=0.5 \text{ cm}$, (b) $z=1 \text{ cm}$, (c) $z=1.5 \text{ cm}$, (d) $z=2 \text{ cm}$. Other parameters as in Fig. 5.8.

Fig. 5.8b shows the Rabi frequencies further inside the amplifier ($\xi=z=1\text{cm}$). At this point, the saturation regime has not yet been reached. The populations remain almost the same as in the amplifier input (cf. Figs. 5.9a and 5.9b), but the fields have increased exponentially.

Around $\xi=z=1.5\text{cm}$, the saturation starts. The Stokes pulse in the second part of the pump pulse has increased almost to its saturation value (Fig. 5.8c). The dynamics of the density matrix elements is no more determined by the pump field alone (Fig. 5.9c), but the Stokes field starts to strongly affect the $|2\rangle \leftrightarrow |3\rangle$ transition. From Figs. 5.8c and 5.9c we see that around the Stokes pulse maximum, stimulated emission from $|2\rangle$ to $|3\rangle$ takes place, so that level $|2\rangle$ decays into level $|3\rangle$ faster than at the rate Γ_2 , and population starts to accumulate in level $|3\rangle$. Pulse compression can be observed by comparison of the pulse widths in Fig. 5.8b and 5.8c.

In the saturation regime, (e.g. $\xi=z=2\text{cm}$, Figs. 5.8d and 5.9d), we observe two important characteristics: the extremely fast decay of the population of level $|2\rangle$ into level $|3\rangle$, due to stimulated emission during the leading edge of the Stokes pulse; and the onset of saturation of the Stokes pulse, with the available populations in level $|2\rangle$ and $|3\rangle$ roughly equalized. This behavior is reminiscent of the steady-state results discussed in Chapter III. The corresponding Stokes and idler Rabi frequencies are shown in Fig. 5.8d. They exhibit very weak maxima at the beginning of the pump pulse (not visible in the figure for the Stokes field), a steep increase to a maximum value, and a slow decay at the end of the pump pulse. The temporal position of this second maximum moves from the second part of the pump pulse towards and beyond the pump pulse maximum as further propagation takes place.

The steep growth of the second maximum is closely related to the fast relaxation of the population ρ_{22} . Therefore, the coherent Stokes pulse is found to be mainly due to laser action rather than to Raman-type processes. The Stokes pulse stimulates the transition $|2\rangle \rightarrow |3\rangle$, which in turn, in a self-consistent way, contributes to the amplification of the Stokes pulse.

The exponential growth and the onset of saturation of Stokes and idler intensities are illustrated in Fig. 5.10, where we show the total (time integrated) generated intensity. We see again a small dip in the idler intensity near the saturation regime, as was obtained for steady-state propagation in Chapter III.

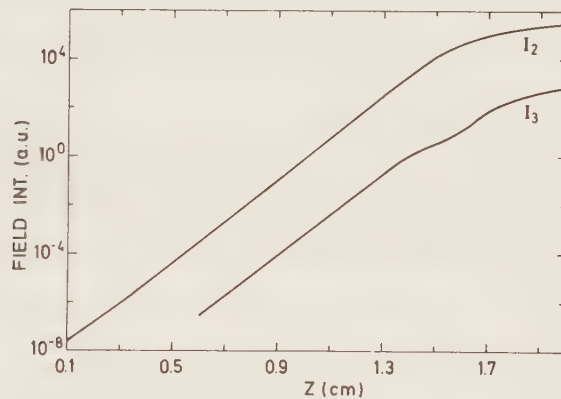


Fig. 5.10

Stokes and idler intensities as function of the amplifier coordinate. The small signal regime, characterized by exponential growth, lasts until $z \sim 1.5$ cm, where the onset of saturation begins.

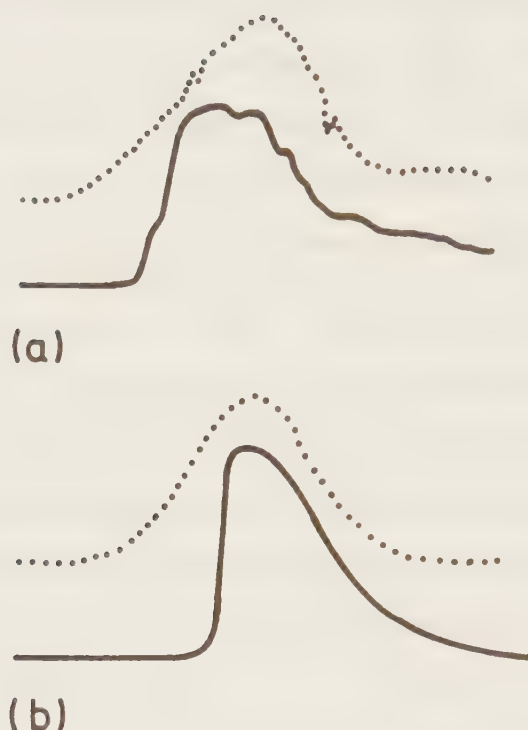


Fig.5.11

Qualitative comparison of (a) experimental and (b) theoretical pulse shapes. The pump pulse in dotted line and the Stokes in solid line. (a) $I_L = 10^8 \text{ W/cm}^2$, $N_2 = 2 \times 10^{13} \text{ cm}^{-2}$, $\Delta_1 = 2.6 \times 10^{11} \text{ Hz}$. (b) $I_L = 3 \times 10^7 \text{ W/cm}^2$, $N_2 = 2 \times 10^{13} \text{ cm}^{-2}$, $\Delta_1 = -0.7 \omega_{21}$, $I_1^{\text{max}} = 6.6 \text{ nsec}^{-1}$, $\gamma_{12} = \gamma_{23} = \gamma_{13} = 10 \text{ nsec}^{-1}$.

A qualitative comparison of the theoretical Stokes pulse shape with experimental results [12] is shown in Fig. 5.11. Our theoretical curve corresponds to an atomic density four orders of magnitude smaller than that of the measurement, and to slower phase relaxation rates. Most characteristics of the experimental shape are nonetheless reproduced: the steep rise, the slower decay, and the position of the maximum at the left side of the pump pulse are in good qualitative agreement. The experimental pulse is however shifted more to the left. This shift is easily understood when one remembers that the Stokes maximum moves to the left as the pulse propagates along the amplifier.

CHAPTER VI

ON SECOND HARMONIC GENERATION IN ATOMIC VAPORS

6.1 Introduction

It is well-known (see e.g./74/) that the second order susceptibility tensor $\chi^{(2)}$ is identically zero in media with inversion symmetry, such as atomic vapors. Thus, within the dipole approximation, second harmonic generation should not occur in such systems. Nevertheless, a number of recent experiments have established in addition to the existence of SHG also its dependence on pump intensity and atomic density. Experimental data are not yet fully understood theoretically. Several explanations have been suggested. Let us briefly describe the most relevant ones.

(1) If neutral atoms only are considered /29/, the electric quadrupole (E2) and the magnetic dipole (M1) interactions, with strengths proportional to the electric field gradient and the magnetic field, respectively, give rise to SHG either directly through a second-order, dipole-forbidden, susceptibility $\chi_{qm}^{(2)}(2\nu_i)$ (*), or through a nonuniform zero-frequency polarization which leads to SHG via $\chi^{(3)}(-2\nu_i; 0, \nu_i, \nu_i)$.

(2) In the presence of photoionization, a process unavoidable when working with high power laser pulses, the free photoelectrons can create net charge densities and quasi-static electric fields E_{DC} leading to SHG via a third order (dipole allowed) susceptibility

(*) The dipole-forbidden second-order susceptibility produces a quadrupole polarization that can radiate when noncollinear, cross polarized beams are applied /20/. Polarization scrambling, or higher order spatial mode patterns in a single beam can lead to quadrupole SHG in a single focused beam /28/.

$\chi^{(3)}(-2\nu; 0, \nu_1, \nu_1)$ /22,29/. The radial motion of the free electrons is governed by the field gradient force (radiation pressure), by the induced restoring electric field forces and by collisions. If the initial kinetic energy of the electron is small, the value of the induced electric field can be roughly estimated as the value required to balance the radiation force. If the initial kinetic energy of the electrons is large, the electrons fly almost freely away from their generation region. In this case a reasonable value for E_{pc} is that for which the electrostatic potential energy equals the initial kinetic energy of the photoelectrons. In most experiments /24-27/, the field gradient forces are negligible in the transport process, which is primarily determined by the outward diffusion of the photoelectrons at the expense of their kinetic energy. Even a very small amount of photoionization turns out to be sufficient to explain the observed SHG intensities.

In this chapter, we concentrate on SHG experiments in Potassium /28/, where a detailed parameter study of second harmonic lines originating from a number of highly excited S, P, and D levels has been performed. (In this thesis we do not consider SHG from P states). We show that the existing dc-field models do not fully satisfactorily predict the experimentally observed dependence of SHG on atomic density and pump laser intensity. As an alternative, we have considered another model where long-range, l-mixing collisions, open a new channel for dipole forbidden transitions to become allowed. This model predicts the correct scaling laws for SHG. However, its application requires the acceptance of a strong assumption which we will discuss later on. Further experiments are thus needed to clarify the situation, and a strong motive for the present studies is to give indications about the direction in which these experiments should proceed. The formalism of Chapter II is suited

for this purpose.

In Section 6.2 we give a survey of the available data, and point to the open questions as regards their theoretical explanation. We then explain briefly in Section 6.3 why collisions should play an important role in atomic SHG. Section 6.4 is devoted to the derivation of the atomic equations of motion in the presence of an interaction mixing opposite parity states. In Sections 6.5 and 6.6, we discuss the photoelectric field model and the model based on collisional mixing, respectively. We show that while the standard dc-field models do not predict the correct scaling of the available data, a "coherent emission assumption" leads to a parameter dependence in reasonable agreement with experiments /27/. A direct comparison is made in Section 6.7, for the cases of S and D lines.

6.2 Comparison of experimental findings with the available theoretical models.

Though popular, predictions based on radial electrostatic fields induced by photoionization are not in full agreement with the experimental results. In Table VI.1 the results of those experiments where parameter dependences of the SHG intensity were measured are summarized. We have also shown attempts which failed to modify the SHG conversion efficiency observed. In the case of resonant pumping (ref. /22,24,26,27/), the dependence of the SHG intensity (I_{SHG}) on atomic density (N) and pump laser intensity (I_L), observed is found to be the same as for ordinary SHG, namely, $I_{SHG} \sim (NI_L)^2$. This scaling is difficult to reconcile with the photoionization model. The radiating polarization is proportional to the atomic density, the two-photon coherence, and the dc-electric field. For an unsaturated two-photon transition and for a

Table VI.1

Atomic vapor	Tl /22/	Na /24/	Li /26/	Na /23/	K /27/
Transition	$6^3P_{3/2} - 7^3P_{3/2}$	$3^2S_{1/2} - 4^2D_{3/2}$	$2^2S_{1/2} - 3^2S_{1/2}$	Nonresonant	$4^2S_{1/2} - n^2S_{1/2}$ $4^2S_{1/2} - n^2D_{3/2,5/2}$
Pulse length	7 nsec	nsec	30 psec pulse train	28 psec	6 nsec
Focal length	40 cm	loosely focused	20,30 cm	46 cm	30 cm
Max.pump intensity	10^7 W/cm^2	10^9 W/cm^2	$2 \times 10^8 \text{ W/cm}^2$	$3 \times 10^{12} \text{ W/cm}^2$	10^8 W/cm^2
Min. num. of photons (f) required for ionization.	3	3	4	5	3
Measured dependences	$N^2 I_L^2$	I_L^2	$N^2 I_L^2$	$N^2 I^5$ $8 > 6 > 1$	$N^2 I_L^2$
Other observa- tions.	Two-photon resonant.	Two-photon resonant. Line shape. Polariza- tion.	Number of ions as function of I_L .		Two-photon resonant. Polariza- tion. Conversion efficiency versus n.
Failed attempts to modify the SHG conversion efficiency.	Magnetic field.	(1) Second laser to monitor ionization. (2) Higher order mode structure in the in- put field. (3) Random polariza- tion modes.	(1) Transverse or longi- tudinal magnetic field up to 60G. (2) Transverse dc-electric field up to 900 V/cm.		

dc-electric field independent of the ion concentration one would indeed obtain the right scaling. The radial electric field strength depends however on the photoelectron density /29/ (the exact form is determined by the transport process), which in turn depends on the atomic density N and on the multiphoton ionization probability $P = Q(\nu_l) I_l^f$. Here $Q(\nu_l)$ denotes the ionization cross section at the laser frequency ν_l and f is the minimum number of laser photons required to reach ionization. This yields a stronger dependence of the SHG on N and I_l than observed. The observed scaling has to be explained by saturation effects /22/; the I_l^2 scaling would be obtained for a saturated two-photon transition and a three-photon ionization such that the dc-electric field would be proportional to the pump intensity. Higher intensity dependences have been observed only for nonresonant pumping /23/.

In an attempt to check the existing models, Freeman et al. /24/ performed a number of experimental tests in Sodium and concluded that none of them could explain their results fully: (1) By using an additional laser to irradiate the vapor, they tried to increase SHG conversion efficiency by increasing the number of ions, but no change was observed. (2) Higher order mode structures in the input laser were introduced as well as polarization scrambling, but the predicted enhancements /20/ were not observed. (3) Polarization measurements showed that the output SHG is not always along the intensity gradients of the input field, nor is it radial, as would be expected from models /23/ and /29/ respectively. These authors also found that the shape of the tuning curve (output power of the SHG as a function of wavelength) is a sensitive function of the vapor density and concluded that the observed SHG arises from a bulk interaction. Similar results for the parameter dependences and polarization of SHG have been obtained by Dinev et al. /27/ in highly excited states (Rydberg states) of

Potassium.

Despite its drawbacks, the dc-field model is still capable of explaining some of the observed features of SHG in atoms, for example the conversion efficiency. Conversion efficiencies up to 10^{-5} - 10^{-6} have been measured in Na /24/ and Zn /25/. These values, of the order of those encountered in allowed three-photon processes, can easily be explained by this model. Jamroz et al. /25/, found for example that 1 to 2% ionization would be enough to explain the measured output power. The corresponding dc-electric fields are about 5 kV/cm. Okada et al. /26/ concluded that the required field in Li vapor should be of the order of 10 to 50 kV/cm, and found experimentally that the application of a dc-electric field up to 900 V/cm had no effect on SHG, consistent with this result.

For completeness, let us note that Freeman et al. /24/ also calculated the power radiated from an isotropic medium by an induced quadrupole moment at the second harmonic frequency, as could possibly be the case for S-D excitation. They obtained an output power 10^{-6} to 10^{-8} times smaller than observed. This rules out electric-dipole forbidden second order susceptibilities as source of the SHG.

This wealth of experimental results, as well as the lack of a satisfying theory to explain them, motivated us to search for alternative symmetry-breaking mechanisms in atomic vapors. In the next Section, we discuss the reasons which lead us to concentrate on l-mixing collisions.

6.3 Why should collisions be considered?

The effect of collisions on line shapes and redistribution of

radiation for dipole allowed transitions has been extensively studied (see e.g. /64/ and references therein). For phenomena such as Raman effect, harmonic frequency generation, frequency sum and difference generation, where coherent excitation plays a central role, it is usually sufficient to treat the collisions within the impact approximation as coherence relaxation processes responsible for line broadening. This picture, which describes adequately the overall effect of strong elastic collisions on allowed transitions, does not require information about the dynamics of the collisions.

The situation is more delicate in the case of forbidden or weakly allowed transitions which are much more sensitive to collisions. Processes such as collision-induced absorption and emission in the weak field regime have been studied both theoretically and experimentally (see e.g. /63/). At high densities and for highly excited states, the oscillator strengths and absorption line shapes depend on the buffer-gas pressure and on the collisionally induced dipole moments (see e.g. /64/). Calculations of these varying dipole moments as function of the internuclear distance are available for alkali-rare gas collisions /65/.

In the case of Rydberg states, collisional l-mixing has been shown to be responsible for reducing the lifetime of the excited state /66/. Measurements and theoretical calculations of l-mixing and broadening cross-sections for alkali-rare gas collisions have also been reported /67,68/. They are of the order of 10^{-13} cm^2 , and always exhibit the same general behavior as a function of the principal quantum number n : for low n they increase as the atomic geometrical size, reach a maximum, and then decrease as the probability of "hitting" the electron decreases. The position of the maximum varies with the collisional partner.

The situation between Rydberg and ground state alkali collisions is

much less clear. Experimental results are available in some cases /69,70/, but the theoretical calculations are rare and not always completely satisfactory. The cross sections are of the order of the geometrical cross sections (10^{-12} - 10^{-11} cm²) and the corresponding collisional mixing can be neglected only when the buffer-gas density is much larger than the alkali density. (*)

Thus, we see that under the conditions under which SHG in atomic vapors has been observed, collisions must certainly be accounted for. They provide the most important line broadening and relaxation mechanism. In addition they provide a l-mixing mechanism, and therefore a channel through which SHG could possibly take place. In the next Section we introduce a formalism allowing to handle a general level mixing scheme. We then specialize first to the case of a dc-field (Section 6.5), and in Section 6.6 to a single model of collisions.

6.4 Two-photon pumping in the presence of a dc-field or l-mixing collisions.

We consider the system of Fig. 6.1, where a strong field pumps a two-photon transition between states $|1\rangle$ and $|2\rangle$ through a set of intermediate states $\{|i\rangle\}$. We assume that one of these $\{|i\rangle\}$ states, labeled $|j\rangle$, is mixed with the upper state $|2\rangle$ e.g. by a dc-field or by collisions. For the sake of simplicity we also assume that the states are nondegenerate. In the semiclassical approximation, the Hamiltonian of the system consists of three terms.

(*) Huge Rydberg-Rydberg spectral broadening for highly excited states ($n > 35$) has also been observed at low pressures /71/; it is mostly due to the Van der Waals interaction between the excited atoms. Under heat-pipe conditions, however, the Rydberg atoms density remains much lower than the ground state alkali density, so that this process can presumably be neglected in our discussion.

$$H = H_a + U + C, \quad (6.1)$$

where, H_a is the unperturbed atomic Hamiltonian, U the pump field-atom interaction as given in Eq. (2.3), and C the additional mixing interaction hamiltonian. For a dc-field E_{dc} , we have $C = -d E_{dc}$; for binary collisions C would be an interatomic potential which depends on the internuclear distance and electronic coordinates. In the first case C is constant whereas in the second it depends on time through the internuclear distance.

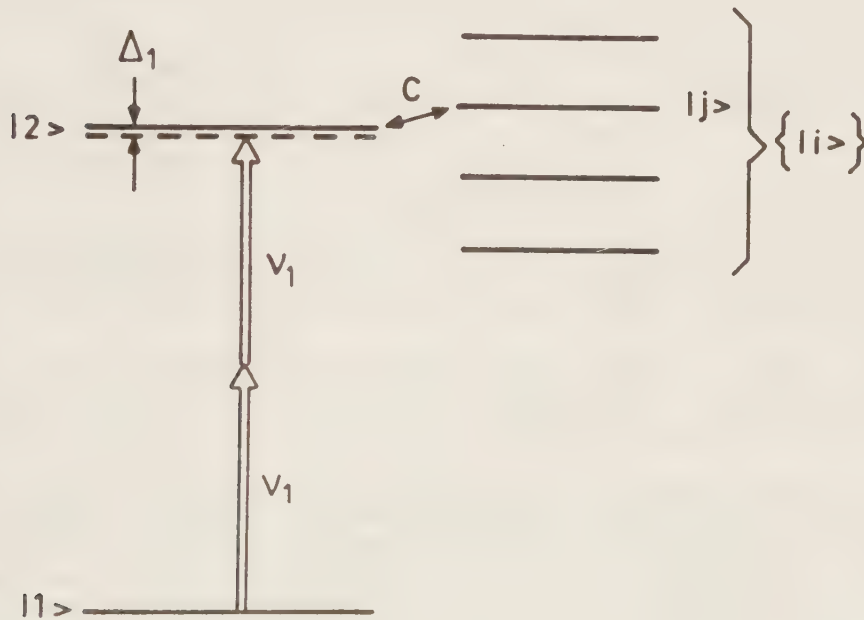


Fig. 6.1

Level scheme showing the strong field of frequency ν_1 pumping the two-photon transition $|1\rangle \leftrightarrow |2\rangle$, and the I -mixing interaction C coupling levels $|2\rangle$ and $|j\rangle$. The set of intermediate states is labeled $\{|i\rangle\}$.

The density matrix equations are obtained as before from the time dependent Schroedinger equation with phenomenological decay constants (see equation (2.4)). A Fourier decomposition similar to that of equation (2.7) leads to the adiabatic elimination of the intermediate states, and to the appearance of the two-photon effective Rabi frequency V_{21} . The only -but central- difference with the results of Chapter II is that now, the nondiagonal matrix element ρ_{ij} contains not only odd harmonics of ν_1 , but also even contributions, in particular $2\nu_1$ (*). Thus the system can now radiate at twice the pump frequency. We obtain :

$$\dot{\rho}_{22} = -\gamma_2 \rho_{22} - (iV_{21} \rho_{12} + \text{c.c.}) \quad (6.2a)$$

$$\dot{\rho}_{11} = (iV_{21} \rho_{12} + \text{c.c.}) \quad (6.2b)$$

$$\begin{aligned} \dot{\rho}_{12} = & i(\Delta_1 + \omega_{s1} I_1 + i\gamma_{12}) \rho_{12} - iV_{12} (\rho_{22} - \rho_{11}) + \\ & i/\hbar \{c_{j2} \rho_{1j} + (c_{22} - c_{11}) \rho_{12}\} \end{aligned} \quad (6.2c)$$

$$\dot{\rho}_{ij} = -i(\omega_{ij} + 2\nu_1 - i\gamma_{ij}) \rho_{ij} + i/\hbar c_{2j} \rho_{12} \quad (6.2d)$$

where $c_{ij} = \langle i | C | j \rangle$ are the mixing interaction matrix elements and all other notations are the same as before. The population of the excited level $|2\rangle$ decays at a rate γ_2 . We neglect the repopulation of the ground level $|1\rangle$ via relaxation processes. The off-diagonal relaxation rates are denoted by γ_{12} and γ_{ij} . Typically, for Rydberg states and heat-pipe conditions, both γ_2 and γ_{12} are collision dominated, but they could also have an intensity dependent contribution due e.g. to photoionization. These processes, which can be described only crudely in terms of simple

(*) The appearance of new frequency components in the presence of collisions was first realized by M. Goeppert-Mayer /43/ in her famous 1931 paper.

decay rates, will not be considered in this chapter. In the density matrix equations we have also ignored the interaction terms due to the second harmonic field, since they are small compared with those involving the pump field.

Equations (6.2a) and (6.2b) for the populations of levels $|2\rangle$ and $|1\rangle$ are the same as obtained with only the pump field present. The mixing interaction can however introduce shifts and broadening into the two-photon coherence ρ_{12} (eq. (6.2c)). Eq. (6.2d) describes the dynamics of the transfer of two-photon coherence ρ_{12} into a coherence ρ_{ij} via the interaction matrix element c_{2j} . It can be solved formally to give

$$\rho_{ij}(t) = \frac{i}{\hbar} \int_0^t dt' c_{2j}(t') \rho_{12}(t') \exp\{-i\Delta_{ij}(t-t')\}. \quad (6.3)$$

where $\Delta_{ij} = \omega_j - \Delta_1 - i\gamma_{ij}$. The dynamics of $\rho_{ij}(t)$ is governed by three different time scales:

- (1) The characteristic time of the mixing interaction. For collisions its scale is given by the collisional time. If the collision partners are Rydberg atoms and weakly excited atoms, it can be estimated by dividing the Rydberg atom diameter by the relative atomic velocity. For the experiment in Potassium /27/ considered here, it is of the order of tenths of nanoseconds. In the case where mixing is due to a dc-field, c_{2j} is a constant (*).

(*) Even if assumed constant in the model, the dc-field as a macroscopic entity takes time to build up. The order of magnitude of this time depends on the photoelectron transport process. If they fly out at expense of their initial kinetic energy, they would leave the pump beam in times of the order of nsec. Another characteristic time scale is given by the inverse of the plasma frequency which depends on electron density and typically may be of the order of psec. If this is the case, a valuable experiment could be done, as suggested by R. Shen /75/ by working with psec-pump pulses: if SHG were due to any macroscopic effect it would eventually disappear for very short pulses. If it were due to microscopic effects it should still be observable.

- (2) The pump dynamics, which determines the evolution of $\rho_{12}(t)$ is characterized by the pump pulse length and the inverse of the two-photon effective Rabi frequency. In our case it is of the order of nanoseconds.
- (3) The "transition time" Δ_{ij}^{-1} between the mixing states. This is the smallest of the three times, provided that the states $|2\rangle$ and $|j\rangle$ are not too close and the effective two-photon Rabi frequency not too high.

In the following, we shall always assume that Δ_{ij}^{-1} is the smallest time, so that Eq. (6.3) can be solved adiabatically to give

$$\rho_{ij}(t) \approx \frac{c_{2j}(t) \rho_{12}(t)}{\hbar \Delta_{ij}} \quad (6.4)$$

In the case where mixing is due to collisions, this approximation implies that real transitions between the states $|2\rangle$ and $|j\rangle$ are neglected.

Since the dipole matrix element d_{ij} is different from zero ($|1\rangle \leftrightarrow |j\rangle$ is a dipole allowed transition), this coherence yields a contribution to the macroscopic polarization which radiates at the second-harmonic frequency $2\nu_1$. This polarization is given by

$$\begin{aligned} P_{SHG} &= N(d_{j1} \rho_{ij} + \text{c.c.}) \\ &= N \left\{ \frac{d_{j1} c_{2j}(t) \rho_{12}(t)}{\hbar \Delta_{ij}} \exp(2i(\nu_1 t - k_1 z)) + \text{c.c.} \right\}, \end{aligned} \quad (6.5)$$

where N is the density of active atoms. From now on, it is convenient to discuss separately the cases of a dc-electric field and of collisions as

possible mixing processes.

6.5 Photoelectric fields

If the mixing process is via a static electric field E_{DC} , the dipole approximation gives $c_{2j} = -d_{2j} \cdot E_{DC}$, and $c_{22} = c_{11} = 0$. Introducing the form (6.4) of the coherence $\rho_{ij}(t)$ into Eq. (6.2c) yields the equation of motion for the two-photon coherence $\rho_{12}(t)$

$$\dot{\rho}_{12} = i(\Delta_1 + \omega_{s1} I_1 + \delta\nu_{DC} + i\gamma_{12}) \rho_{12} - iV_{12}(\rho_{22} - \rho_{11}) \quad (6.6a)$$

where the dc-Stark shift $\delta\nu_{DC}$ is given by

$$\delta\nu_{DC} = \frac{|d_{2j} \cdot E_{DC}|^2}{\hbar^2 \Delta_{ij}} \quad (6.6b)$$

The total energy W_{SHG} radiated at the frequency $2\nu_1$ is obtained by integrating the equation for the second harmonic field, with the source polarization P_{SHG} . This gives with Eqs. (6.6a) and (6.5),

$$W_{SHG} = \frac{4\nu_1^2 N^2 |d_{ij}|^2}{c \epsilon_0} \int_{-\infty}^{\infty} dt \left| \int_0^L dz \rho_{21}(z, t) F(z, t) \exp(i(2k_1 - k_{SHG})z) \right|^2. \quad (*) \quad (6.7)$$

where L is the length of the sample, and we have introduced the "mixing function"

$$F(z, t) = - (d_{2j} \cdot E_{DC}) / \hbar \Delta_{ij} \quad (6.7a)$$

$$(*) \quad \rho_{21} = \rho_{12}^*, \quad \Delta_{ij} = \Delta_{ji}^*, \quad c_{2j} = c_{j2}^*, \quad \gamma_{21} = \gamma_{12}^*.$$

The behavior of the radiated energy (6.7) is determined essentially by three quantities: (1) the two-photon coherence $\rho_{21}(z,t)$ which can be assumed independent of z if pump depletion and other propagation effects are ignored; (2) The phase mismatch function $\exp(i(2k_1 - k_{SHG})z)$, where k_{SHG} is the second harmonic wave vector in the medium; (3) the mixing factor $F(z,t)$ which contains the effects due to the photoionization-induced dc-field. We proceed now to discuss each contribution separately.

6.5.1. Two-photon pumping

The evolution of the two-photon coherence $\rho_{21}(z,t)$ is determined by Eq. (6.6a). In order to solve it, one needs to know the effective Rabi frequency V_{21} , the dynamic and static Stark shifts $\omega_{S1}I_1$ and δV_{DC} , as well as the relaxation rate γ_{21} . The effective flipping rate V_{21} and the ac- and dc-Stark shifts are determined by dipole matrix elements. For Rydberg states these can be evaluated by the Bates-Daamgard approach /72/ using Coulomb wave functions including the quantum defect. This approach gives the analytical value of the dipole moments in the form of an alternating series. For high principal quantum numbers, however, this series converges very slowly, and we found it more convenient to perform the radial integrals numerically. Our computational results turn out to be more accurate for dipole moments between neighbouring states than for those between ground state and highly excited Rydberg states.

For pressure-broadened media, the off-diagonal relaxation rate γ_{21} is calculated from

$$\gamma_{21} = \frac{1}{2} (\gamma_2 + \gamma_1) + \gamma_{ph}$$

where the phase mixing contribution due to collisions can be estimated from $\gamma_{ph} = \langle N \sigma v \rangle$; σ is the collisional cross section and v the relative velocity. For a Maxwellian velocity distribution and considering both K-K and Ar-K collisions we get

$$\gamma_2 = \frac{\gamma_2}{2} + \left(\frac{16k_B T}{\pi m_K} \right)^{1/2} \left(\sigma_{KK} N_K + \sigma_{KA} N_A \left(1/2 \left(1 + \frac{m_K}{m_A} \right) \right)^{1/2} \right) \quad (6.8)$$

where N_K is the Potassium density, m_K the mass of the Potassium atom, and T the temperature inside the heat pipe. (Corresponding buffer-gas quantities are indexed by A). N_K is evaluated from the temperature using the empirical fit given in Ref./6/; N_A is obtained from the measured buffer gas pressure. Potassium-rare gas collision broadening cross sections σ_{KA} have been calculated according to /67/; this model is in good agreement with experiments in Cs and Rb and is probably also reasonable for Potassium. For K-K* collisions the situation is less clear, and we have opted by using the geometrical cross-sections. Fortunately we often have $N_K \ll N_A$ which implies the dominance of rare gas-Potassium collisions.

Difficulties are also encountered in estimating the population decay rate γ_2 which has the same functional form as equation (6.8), but where the collisional cross sections are those for population depletion. (The broadening is mainly due to elastic collisions, whereas depletion is due to inelastic ones). The spontaneous decay of state $|2\rangle$ is already negligible at intermediate pressures and the upper state lifetime is determined by collisional l-mixing /66/.

Equations (6.2a), (6.2b) and (6.6a) can be solved numerically for Gaussian pulses. The inclusion of a transverse pump beam profile is straightforward, provided that beam curvature, self-focusing, and

defocusing effects are neglected. The total radiated energy W_{SHG} is then obtained by integrating Eq. (6.7) over the transverse beam profile. We consider a Gaussian incident pump pulse of the form

$$I_L(r_\perp, t) = I_{L0} \exp\left(-\frac{r_\perp^2}{r_0^2} - \frac{t^2}{\tau_L^2}\right). \quad (6.9a)$$

where $r_\perp$ is the transverse coordinate. The peak intensity I_{L0} is given in terms of the pulse energy W_L by

$$I_{L0} = \frac{W_L}{\pi^{3/2} r_0^2 \tau_L} \quad (6.9b)$$

and r_0 and τ_L are the $1/e$ radius and temporal width of the laser pulse respectively. In the experiment /27/ $\tau_L \sim 3\text{ns}$ and $r_0 \sim 0.1\text{mm}$.

Drastic simplifications follow if the off-diagonal relaxation is much faster than the effective two-photon Rabi frequency and the inverse pulse duration: $\gamma_{12} \gg V_{21}, \tau_L^{-1}$. In this case, ρ_{12} follows adiabatically the pump pulse. If furthermore the pump field is non saturating, i.e., $|V_{21}|^2 T_1 / \gamma_{21} \ll 1$, we obtain the simple result

$$\rho_{21} = -iV_{21} / (\gamma_{21} + i\Delta_1' + i\delta\nu_{DC}). \quad (6.10)$$

Whether two-photon saturation is reached or not for a given pump intensity depends on the values of the relaxation rates γ_2 and γ_{21} . If the upper state were not depleted by collisions, saturation would readily be achieved. This is however, not the case to be expected under the experimental conditions of ref. /27/.

For square pulses, the ac-Stark shift included in eq. (6.10) can obviously readily be canceled by a proper choice of pump frequency. In the case of Gaussian pulses, we have shown in Chapter V that the

resonance condition can be roughly achieved by detuned pumping. We therefore assume here that $\Delta_1' = 0$, and also neglect the dc-shift compared to γ_{21} , so that the temporal and spatial dependences in (6.10) come only from V_{21} .

6.5.2. Phase mismatch factor

In the last section we saw that the two-photon coherence is independent of z if pump depletion and other propagation effects are ignored. If the mixing factor $F(z, t)$ is independent of z , as in the case of an uniform static field E_{DC} , then the integral over z in Eq. (6.7) reduces to the integral of the phase mismatch factor, namely

$$\int_0^l dz \exp(i\Delta k z) = l^2 \frac{\sin^2 \frac{\Delta k l}{2}}{\left(\frac{\Delta k l}{2}\right)^2} \quad (6.11)$$

where $\Delta k = 2k_1 - k_{SHG}$. The $(\sin x/x)$ dependence is typical for plane wave propagation /6/. For focused beams the dependence can be completely different /6/. We have also seen in Chapter III that for exponentially growing fields, the effect of phase mismatch can be negligible.

The phase mismatch Δk was calculated by means of the indices of refraction for Potassium, as defined in Chapter II:

$$\Delta k = \frac{2\nu_1}{c} \{n(\nu_1) - n(2\nu_1)\} \quad (6.12)$$

For the buffer-gas dispersion, Argon in our case, we have used the results of Ref./73/. The Potassium dispersion predominates, and the $\Delta k l/2$ factor must be already considered for Potassium densities larger than 10^{15} cm^{-3} .

6.5.3. DC-field mixing factor

If the static electric field E_{DC} is a constant, independent of z, t , and $r_{\perp}$, then the integral (6.7) reduces to

$$W_{SHG} = \frac{4\nu_i^2 N_K^2 |d_{ij}|^2}{c\epsilon_0} \frac{|d_{2j} \cdot E_{DC}|^2}{(\hbar \Delta_{ij})^2} l^2 \frac{\sin^2 \frac{\Delta k l}{2}}{\left(\frac{\Delta k l}{2}\right)^2} \iint dr_{\perp} dt |g_{2i}|^2 \quad (6.13)$$

This result shows that if E_{DC} does not depend on the pump intensity, nor on the atomic density, then the measured scaling of SHG intensity with Potassium density and pump laser intensity $I_{SHG} \sim (N_K I_L)^2$ could be reproduced. Furthermore, the measured conversion efficiencies would be obtained, e.g. for the 13S level, with $E_{DC} \sim 120 \text{ V/cm}$. The corresponding dc-Stark shift ($2.5 \times 10^8 \text{ sec}^{-1}$) is negligible.

The temporal behavior of E_{DC} depends on electron density oscillations and ionization dynamics, damping conditions, pump beam focusing and electron transport. Let us assume that three-photon ionization takes place at a rate

$$\dot{N}_i = Q(\nu_i) N_K I_L^3 \quad (6.14)$$

and that the electrons diffuse out of the beam region at the expense of their initial kinetic energy. In the low K -density limit, and for electrons with an initial energy of 1eV, the restoring forces due to E_{DC} are negligible. The electron excursion is found to be of the same order of magnitude as the beam radius, but considerably less than the free-flight distance because of the collisions with buffer-gas atoms. The average E_{DC} field turns out to be proportional to the ion density, which in turn is proportional to the Potassium density and to the third

power of the intensity. At high atomic densities, the action of the E_{dc} field on the electrons can no more be neglected, and the ion density dependence is expected to turn gradually to $N_i^{1/2}$. It has also been estimated that, under the experimental conditions of Ref. /27/, an ionization degree of the order of 10^{-6} would be sufficient to produce the required dc-field.

Summarizing: for low ion densities $\langle E_{dc} \rangle \sim Q(\nu_i) N_K I_L^3$, and $W_{SHG} \sim N_K^4 I_L^8$; for higher ion densities $\langle E_{dc} \rangle \sim N_K^{1/2} I_L^{3/2}$, and $W_{SHG} \sim N_K^3 I_L^5$. This, assuming that the predominant contribution to the phase relaxation rate γ_{21} (Eq. 6.8) is due to collisions with buffer-gas atoms, and that no two-photon saturation is reached.

These predictions are hardly reconcilable with the experimental results. Weaker dependence of SHG on the pump intensity could be achieved if the two-photon transition were saturated. In this case, the two-photon coherence ρ_{21} would be pump-independent, and the ionization rate would then scale as I_L /76/, so that W_{SHG} would be proportional to I_L^2 (in the low ion density regime). This would however imply that for low pump intensities stronger than squared intensity dependence should be observed, in disagreement with experimental results (see Table VI.1). Thus even though this model is able to predict the measured efficiencies correctly, it is difficult to reconcile it with the other features of SHG. In particular it is hard to understand why the photoionization induced dc-field should be independent of the ion density and the pump intensity.

6.6 Collisional mixing

Let us now return to equation (6.5) for the polarization at frequency

$2\nu_1$, and discuss what would happen if the mixing of the states $|2\rangle$ and $|j\rangle$ were due to collisions. In this case, the coefficient $c_{2j}(t)$ varies within the collisional time scale, and the expectation value of the dipole operator oscillates at $2\nu_1$, but with a time dependent amplitude $c_{2j}(t)$. Physically, this means that during a collision, a channel giving a time dependent dipole moment radiating at $2\nu_1$ is opened.

This picture can be better understood by considering the various time scales appearing in Eq. (6.5): $\rho_{12}(t)$ oscillates at the two-photon effective Rabi frequency $|V_{21}| \sim 10^9$ Hz. $c_{2j}(t)$ varies in the collisional time scale $\tau_c \sim n^2/v \sim 10^{-10}$ sec, and $2\nu_1 \sim 10^{15} - 10^{16}$ Hz. Thus $|V_{21}| < \tau_c^{-1} \ll 2\nu_1$, and the two-photon coherence $\rho_{12}(t)$ is slowly varying compared to $c_{2j}(t)$ (i.e., during a collision).

Since the macroscopic dipole moment (6.5) depends on the collisional parameters through $c_{2j}(t)$, to obtain the radiated intensity a collisional average must be performed. It is important at this point to remember that SHG is a coherent effect in the sense that the emitted radiation has a reference phase imposed by the coherent pumping. But within a slice of medium one wavelength long, there are about 10^7 alkali atoms undergoing collisions. If only close range collisions are relevant in mixing the states, the collisional phase shifts, which are strongly dependent on the impact parameter, will blur this reference phase, so that no coherent build up of the SHG can be expected. The situation changes in the case of long range collisions with impact parameter larger than the Weisskopf radius (see e.g. /78/); in this regime, the collisional phase shifts do not vary strongly with the collisional parameters, and radiation emitted in similar kind of collisions could add coherently. To obtain the energy radiated by the sample in a time interval between t_0 and $t + \Delta t_0$, with $\Delta t_0 \gg \tau_c$, we solve the field equation for the Fourier transformed field amplitudes in the slowly

varying approximation:

$$\mathcal{E}_\omega = \frac{2i\nu_1}{c\epsilon_0} \int_0^l dz \rho_\omega(z, t_0) \exp(i(2k_1 - k_{SHG})z). \quad (6.17)$$

To calculate

$$\rho_\omega(z, t_0) = N_k \rho_{21}(z, t_0) \overline{c_{j2}(b, \underline{v}, \omega)} \frac{\underline{d}_{ij}}{\hbar \Delta_{j1}} \quad (6.18)$$

We have taken into account the fact that $\rho_{21}(z, t)$ varies slowly during a collision, so that we can remove it from the Fourier integral and evaluate it at the time t_0 when the collision takes place. In (6.18) we have introduced explicitly the dependence on the relative atomic velocity $\underline{v}$ and the impact parameter b . $c_{j2}(b, \underline{v}, \omega)$ is the Fourier transform of $c_{j2}(b, \underline{v}, R(t))$, where $R(t)$ is the internuclear coordinate. The bar means collisional average.

If we neglect pump depletion, the energy radiated in Δt_0 is

$$\int_{-\infty}^{\infty} d\omega |\mathcal{E}_\omega| = \frac{4\nu_1^2}{c^2 \epsilon_0^2} N_k^2 |\rho_{21}(t_0)|^2 \int_0^l dz' \int_0^l dz'' \exp(i\Delta k(z' - z'')) \int_{-\infty}^{\infty} d\omega |\overline{d_{12}}|^2 \quad (6.19)$$

where we have introduced the quantity d_{12} which can be interpreted as the mean value of the collisionally induced dipole moment

$$\overline{d_{12}(\omega)} = \frac{\underline{d}_{ij}}{\hbar \Delta_{j1}} \overline{c_{j2}(b, \underline{v}, \omega)} \quad (6.20)$$

If we assume that the phase of the dipole moment d_{12} is not strongly perturbed during a collision, we can consider its magnitude only and substitute the average dipole moment squared by the average of the

squared dipole moment: $|\overline{d_{12}}|^2 \rightarrow |\overline{d_{12}}|^2$. This approximation, although difficult to justify has already been used in /77/ in their analysis of gain enhancement by collisions. Here, we use this approximation without further justification, and study what its consequences would be for SHG in atomic vapor. The expression $|\overline{d_{12}}|^2$ appearing in the Eq. (6.19) for the radiated energy is then

$$|\overline{d_{12}}|^2 = \left| \frac{d_{ij}}{\hbar \Delta_{ji}} \right|^2 \overline{|c_{j2}(b, y, \omega)|^2} \quad (6.21)$$

Our assumption means that atoms radiate collectively with an effective dipole moment given by the square root of Eq. (6.21).

6.6.1. A crude collisional model

Most of the treatments /67,68/ of the interaction between Rydberg atoms and neutral perturbers are based on the fact that the Rydberg atom is much larger than the perturber, and the electron is loosely bound to the Rydberg core. The problem can then be reduced to the highly localized electron-perturber interaction expressed by the Fermi contact potential

$$C = 2\pi L \delta(\underline{r} - \underline{R}) \quad (6.22)$$

where L is the scattering length obtainable from electron-perturber scattering data. $\underline{R}$ is the position of the perturber respect to the Rydberg core and $\underline{r}$ is the electron's coordinate (see fig. 6.2). The perturber is regarded as a structureless particle following the straight-line trajectory

$$\vec{R} = \vec{b} + \vec{v} t \quad (6.23)$$

where $\vec{b}$ is the impact parameter and $\vec{v}$ the relative velocity.

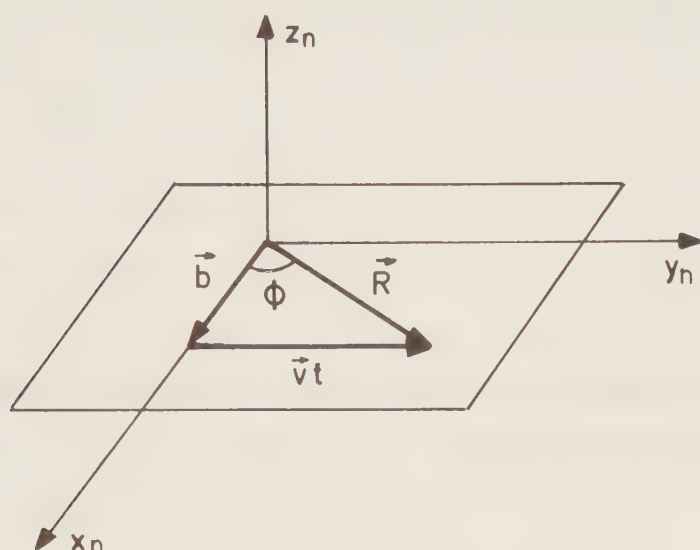


Fig. 6.2

The collision frame: The internuclear vector $\vec{R}$ follows a straight line trajectory with impact parameter $\vec{b}$ and velocity $\vec{v}$. The z axis is chosen perpendicular to the collisional plane defined by $\vec{v}$ and $\vec{b}$.

The model can be improved /67/ by adding to the Fermi potential the interaction potential between the neutral perturber and the Rydberg core. If the perturber has no permanent dipole or quadrupole moment, the leading term in the long range electrostatic interaction is $-\alpha/2R^4$, where α is the polarizability of the perturber. This interaction is much less efficient in mixing the Rydberg states than the Fermi interaction and is rather responsible for collisional shifts and broadening. To calculate the interaction matrix elements for the Fermi potential is straightforward:

$$c_{ij}(R(t)) = 2\pi L \psi_i(R(t)) \psi_j(R(t)) \quad (6.24)$$

The collisional matrix elements were calculated in a fixed frame with the quantization axis z_n perpendicular to the collisional plane. It is a natural choice for l-mixing collisions because angular momentum can only be transferred perpendicular to the scattering plane, which is defined by $x_n || \mathbf{b}$ and $y_n || \mathbf{v}$ (see fig. 6.2). In this plane the wave function takes the form

$$\psi_{nlm}(R) = R_{nl}(R) Y_{lm}(\pi/2, \phi). \quad (6.25)$$

where $\tan \phi = vt/b$. For the radial part $R_{nl}(r)$, we have solved numerically the differential equation for the outer electron in the Coulomb potential of the core including quantum defect corrections.

The transformation from this collision frame to the laboratory frame is accomplished by a rotation through the Euler angles α, β, γ , which determine the orientation of the collision frame in space:

$$Y_{lm}(R) = \sum_{\mu} Y_{l\mu}(\pi/2, \phi) D_{\mu m}^l(\alpha, \beta, \gamma) \quad (6.26)$$

$D_{\mu m}^l(\alpha, \beta, \gamma)$ are rotation matrices.

In evaluating (6.21) we must average over all orientations of the collision frame, as well as over impact parameter and velocity. For not very high densities the time T between collisions is larger than the collision time τ_c , $T > \tau_c$, and we can work within the binary approximation. Furthermore, $T \ll |V_{21}|^{-1}$ and an average over collisions during Δt_c , $T < \Delta t_c < |V_{21}|^{-1}$ can be done. The number of collisions per atom during Δt_c , with impact parameter between b and $b+db$ and velocity between v and $v+dv$ is $N_A \int 2\pi b db v f(v) dv \sin \beta d\beta d\alpha d\gamma \Delta t_c$, where $f(v)$ is

the velocity distribution function. If the mixing is assumed independent on v , we can just introduce the most probable velocity $\bar{v} = \int dv v \mathcal{H}(v)$, so that (6.21) reads

$$\overline{|d_{12}|^2} = \left| \frac{d_{ij}}{\hbar \Delta_{ji}} \right|^2 N_A \Delta t_0 2\pi \bar{v} \int_{b_w}^{\infty} b db \int_0^{\pi} \sin \beta d\beta \int_0^{2\pi} d\alpha \int_0^{2\pi} d\delta |c_{j2}(b, v, \omega, \alpha, \beta, \delta)|^2 \quad (6.27)$$

The integral of $\overline{|d_{12}|^2}$ over ω in (6.19) is equivalent to the integral over time of its inverse Fourier transform. This integral was done numerically for different impact parameters, and then integrated over impact parameters with an spline fit. In order to include only soft collisions, the integral should be done from the Weisskopf radius to infinity. From a semiclassical approximation by Omont /67/, we can calculate the Weisskopf radius

$$b_w = \frac{1}{2} \left(\frac{0.53 L}{n^3 \bar{v}} \right)^2 \quad (6.28)$$

For potassium-argon collisions $L = -1.7$, so that for $n=10$ and temperatures around the 470°C , $b_w \sim 40$ a.u., whereas the wave functions have their maximum around $n^2 a_0 \sim 100$ a.u., which yields high probability of mixing for large impact parameters, beyond the Weisskopf radius. The total energy radiated during the pump pulse is obtained by integrating (6.19) over t_0 (coarsened grained average). The results are compared with the experiment in the next section.

6.7 Comparison with the experiment

Here we give the results obtained by integrating Eq. (6.19) over t_0

to obtain the total second harmonic energy. In fig. (6.3) we give both the experimental data and theoretical fit for I_{SH6} as a function of the input intensity. The functional dependence is quadratic; in the theoretical curve some saturation is observed unlike in the experimental data. The discrepancy probably indicates that the upper state $|2\rangle$ is collisionally depleted at a higher rate than assumed in the theoretical calculations.

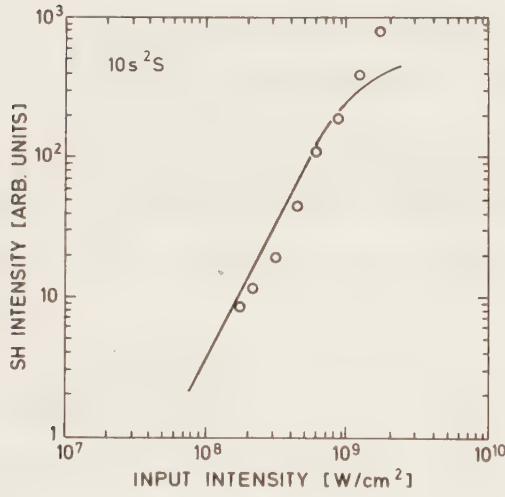


Fig. 6.3

SHG intensity as function of the pump intensity. The dots are experimental data from Ref. /27/, and the full line is the theoretical curve.

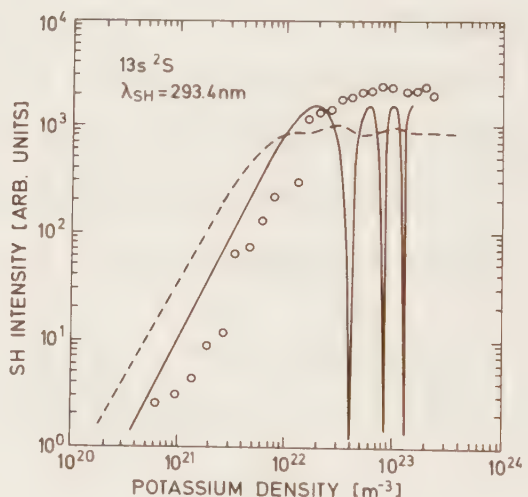


Fig. 6.4

SHG intensity as function of the Potassium density. The dots are the experimental results of Ref. /27/. Full line: Theoretical curve including phase mismatch for a plane wave. Dashed line: theoretical curve including density variations.

The second harmonic intensity vs. the Potassium density is shown in Fig. 6.4. The points are the experimental data, whereas the full and dashed lines are theoretical fits. The theoretical curves are calculated including phase mismatch effects which become important for Potassium densities around $10^{15} - 10^{16} \text{ cm}^{-3}$. The full curve corresponds to the case

where no density fluctuations are assumed. We observe narrow minima, with increasing period, due to the phase mismatch factor. In the heat pipe, however, many inhomogeneities can appear, especially when varying the atomic density by changing the temperature. We would expect fluctuations in the atomic density within the length of the interaction region and also within the coherence length. Fluctuations in the coherence length can arise not only from density fluctuations but also from variations of the index of refraction due to the two-photon pumping. We have simulated density fluctuations up to 30% and obtained the dashed curve, where the phase mismatch minima are averaged out and the experimental plateau reproduced. We should however remember that a different kind of phase mismatch function would be obtained if the plane wave approximation is relaxed, and this fact may also be the reason for the absence of minima in the experiment.

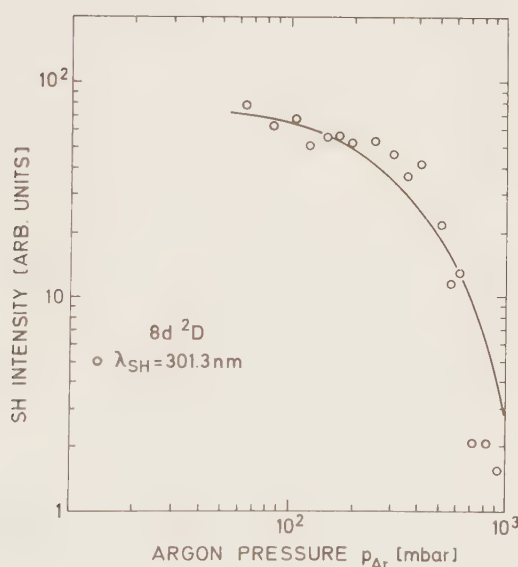


Fig. 6.5

SHG intensity as function of the buffer-gas pressure. The dots are experimental data of Ref. /27/ and the full curve is the theoretical result.

Fig. 6.5 shows the experimental data and theoretical fit for the I_{SHG} dependence on the buffer-gas (Argon) pressure. The curvature of the curve depends on the pump intensity.

All theoretical curves show acceptable agreement with the experimental data, which however by no means is a sufficient proof of the validity of the model. The same curves, except that for the dependence on buffer-gas pressure would be obtained within the photoionization induced dc-field model if the field is assumed independent on the pump intensity and atomic density.

CHAPTER VII

CONCLUSIONS AND OPEN QUESTIONS FOR FUTURE RESEARCH

Within the framework of semiclassical theory where matter is described quantum mechanically and fields classically we have studied several nonlinear processes taking place in two-photon pumped systems. The matter-field equations have been solved in a self-consistent way with the polarization created by the field in the medium constituting a source for the field.

The solution of the semiclassical matter-field equations for a particular problem is by no means a straightforward task even for two-level systems, and the complexity increases with the number of fields and active levels involved in the problem. A major development was achieved here by extending the two-photon two-level model to a three-level system pumped by a two-photon transition. This model has enabled us to go beyond the predictions of perturbation theory and to study analytically several aspects of SHRS and the related four-wave mixing process. Two-photon pumping constitutes a common feature to all applications, including the analysis of several parameter dependences in SHG. The formalism is in general suited to describe two-photon resonant processes.

In the second Chapter we have outlined the procedure applied and established the conditions under which the model is valid. The formalism is based on the distinction between resonant and nonresonant interactions. This distinction makes possible the adiabatic elimination of nonresonant contributions in the matter equations and a consequent reduction of the atomic model to a simple three-level system.

Furthermore we have also succeeded in separating the resonant and nonresonant contributions in the field equations: while the resonant contribution is responsible for either amplification or absorption, the nonresonant contributions give rise to a linear polarization whose effect on the field propagation can be described by an index of refraction and a weak background absorption. If such a separation is not performed, the resonant interaction would enter into the field propagation equations as a resonant denominator, which without the inclusion of a line broadening or a laser width diverges at resonance. By this procedure we avoid the calculation of resonant susceptibilities, and consider the effect of the resonant interaction in a much cleaner way.

We have first solved the matter-field equations in steady-state, to first order in Stokes and idler fields, and recovered the exponential growth of SHRS predicted by perturbation theory. We have also studied the growth of the idler field and found that it is closely related to that of the Stokes field: in the small gain regime both fields grow exponentially with the same gain.

Beyond the small gain region, experimental results exhibit a linear dependence of the hyper-Raman line on the optical depth, which is not predicted by higher order perturbation theory. We have shown, by solving the matter equations to second order in the Stokes field, that this saturation is not directly related to the two-photon saturation, but to the interaction of the Stokes field with its resonant transition. (In this context the two-photon pumping populates the upper level with the Stokes field tending to equalize the population in levels $|2\rangle$ and $|3\rangle$). The pump intensity, however, affects the Stokes field saturation (see Eq. (3.16)). The fact that the saturation region is easily reached in a short optical depth is due to the large exponential Stokes gain, and

clearly shows the inadequacy of perturbation theory, as well as of theories which neglect the population in the upper state to describe the experimental findings.

The saturation of the idler field has not been studied analytically because of the intricacy of the equations involved. A numerical solution shows two interesting features: the appearance of a dip in the intensity close to the point where saturation sets in (pumping at resonance) and the absence of oscillations due to phase mismatch effects (masked by high gain). The dip is due to a change of sign in the Rabi frequency and corresponds to the cross over from the small gain region where a pure four-photon process is responsible for both Stokes and idler generation to the region where population starts to accumulate in the upper levels and where the Stokes field interacts strongly with its resonant transition.

Another interesting point discussed was the effect of the phase mismatch on the idler field propagation. The well-known phase-mismatch effects in Second and Third Harmonic Generation show an oscillatory behavior. However we found that no oscillations appear in the idler output intensity, at least under the experimental conditions analyzed. The phase mismatch actually reduces the output intensity in a similar way as in SHG and THG but due to the exponential growth of the fields we do not obtain a simple $(\sin x/x)^2$ functional dependence.

Future theoretical research in this field could include the treatment of $(2\nu_1 + \nu_2)$ sum frequency generation which has also been studied experimentally /12/, as well as applying the formalism to study the Stokes and idler generation in Rydberg atoms, which possibly show different behavior. The description used here was adapted to a three-level system, where the third level relaxes much quicker than the

upper one. For Rydberg states the difference in relaxation rates is not so large, so that saturation of the hyper-Raman transition can occur quickly. Also the phase mismatch increases and therefore it could be possible that the output idler intensity is strongly reduced.

From the point of view of the dynamics of this three-level system it would also be of interest to relax the condition we imposed to give energy conservation, namely $2\nu_1 = \nu_2 + \nu_3$, and study the system's response to three externally applied fields, a pump and two probes, where new interesting effects due to interferences and beat frequencies are to be expected.

Our study of the transient behavior opens up new possibilities to measure Rabi frequencies and decay rates, especially those connected with the two-photon coherence between states of the same parity. The time-frequency method employed here, provides a means of 'seeing' the evolution of Stark shifts and splittings. We have assumed $\gamma_2 < \gamma_{21} < \gamma_{23}, \gamma_{13}$, (reasonable at low pressures when $\delta_2 < \delta_3$), and studied the optical nutation and "free induction decay" signals. We found that Raman-type processes have their maximum in the optical nutation regime and quickly die out when the pump field is switched off. The "free induction decay" signal shows a dominant population-type line. A strong signal reaching its maximum at a delayed time with respect to the switching off of the pump field was found. This interesting feature is due to the coupling of the upper state population with the Stokes probe field.

A problem not discussed here should be mentioned as a topic for future research. It is known that the Raman line dies out as soon as the pump field is switched off, i.e., as soon as the two-photon coherence dies out. There is however the possibility of applying a second probe at

the idler frequency ν_3 and looking at either the idler or at the Stokes gain spectrum after the laser is switched off. In the same way as the interaction with the Stokes field brings into play the population of the upper state at the end of the pulse, the application of the idler probe "revives" a two-photon coherence which gives rise to a new signal at the position of the Raman line in the "free induction decay" regime. This signal has an additional feature: its shape depends on the relative phase of pump, Stokes and idler fields, and can therefore constitute a valuable tool for measuring these phases.

The study of the Stokes gain for Gaussian pulses, as well as of pulse propagation through the medium gives us additional information on the temporal behavior of SHRS and idler field with respect to the pump field. For pumping at resonance and with slow decay constants both the optical nutation and the population transient in the "free induction decay" regime contribute to the Stokes gain.

A clear temporal distinction between population- and Raman-type lines was seen for large pump detunings. While the Raman line exists only during the application of the pump pulse, after the pulse has passed there appears a strong population-type line with a long tail arising from the slow population decay of the upper state.

We have obtained a computer simulation, under experimental conditions, of pulse propagation in a heat-pipe oven for the Stokes and idler fields. To hold the computer time within reasonable limits, we were forced to work at lower atomic densities than those used in the experiment. The Stokes field shows two maxima as observed in the experiment, one before the pump pulse maximum, mainly of Raman character, and a second corresponding to the population transient which slowly moves towards the pump maximum as it propagates through the

medium.

The last topic we considered is SHG in gases. After an analysis of the relevant available models, we considered in some detail two mechanisms: the photoionization induced dc-field model and a collisional model put forward in this work. We found that the proposed electric field model failed to explain the observed experimental dependences if one considers the dependence of the dc-field itself on the pump intensity and atomic density. If for some reason the dc-field depends on none of them, or if the two-photon transition is saturated, so that the dc-field is proportional only to the pump intensity, and is assumed to be independent of the atomic density, the dc-field model gives the correct dependences.

On the other hand the collisional model reproduces the experimental dependences, but relies on the assumption that the atoms radiate in a collective way with a mean square collisionally induced dipole moment. This situation is expected to occur in connection with soft collisions which are able to transfer two-photon coherence to a pair of states connected by a radiative transition, without introducing large collisional shifts. In allowed transitions strong collisions mainly introduce large phase shifts, destroy coherences, broaden and alter line shapes (especially at the line wings), and deplete the atomic levels by transferring population to neighboring states; all of this happening in times much shorter than the time constants involved in the dynamics of field-matter interaction. Here we have inspected the behavior "inside" the collision, where forbidden radiative processes can become allowed. Our model should be further tested experimentally and the theoretical description refined within the framework of collisionally induced processes. The study of such processes (and laser-induced collisions) already constitutes a major field in physics, but still offers a great

variety of open questions.

As a consequence of our theoretical analysis we realized the need for new experimental measurements of SHG under more controlled experimental conditions. The experimental evidence so far available has been obtained by working with single heat-pipes. Their main advantage is that the pressure of the metal vapor inside the heat-pipe oven can be measured directly from the pressure of the confining inert gas without relying on vapor pressure curves. The heat-pipe regime is achieved when the metal vapor pressure equals that of the inert gas. In this regime a zone of homogeneous pure metal vapor of well defined pressure is confined by the inert gas lying close to the windows.

In some experiments, however, no advantage has been taken of these characteristics: (1) Higher buffer-gas pressures than the partial metal vapor pressure have been used (e.g. /22,23,27/); therefore the heat-pipe does not work in the heat-pipe regime, i.e., instead of having a well defined metal vapor zone, a mixture of both gases is obtained along the heated region. The density is then difficult to control and is not homogeneous. (2) The SHG intensity dependence on atomic density has been studied by varying the metal vapor temperature /22,27/; therefore the vapor pressure curves were needed, transforming small errors in the temperature measurement into large errors in the pressure measurement. (3) The absence of a well defined metal vapor zone leads also to difficulties in the determination of the interaction length. (4) An increase of the metal vapor pressure at constant buffer-gas pressure leads also to a decrease of the partial buffer-gas pressure in the heated region, and possibly to changes in the phase relaxation rates. Other difficulties related to the use of heat-pipes are the following: (5) Relatively high atomic densities ($N \sim 10^{15} \text{ cm}^{-3}$) are needed, so that phase-mismatch effects become important. In addition, the density

gradients in the transition zones in the heat-pipe have to be considered. For focused beams of high intensity theoretical considerations of these effects are complicated. (6) In the case of Potassium /27/, the presence of strong parametric lines which overlap the weak SHG signal makes difficult its detection.

To overcome some of these problems and obtain more reliable data and further information we suggest new experiments with atomic beams where the complexity of the system, at least from the theoretical point of view, is strongly reduced: (1) The buffer-gas is absent. (2) Collisions are strongly reduced. (3) Because of the reduction of collisional rates, the phase relaxation rates are reduced, so that two-photon saturation can be easily achieved even at moderate pump intensities. (4) It is possible to work at low atomic densities, where phase-mismatch effects can be neglected and the parametric lines are strongly reduced, facilitating SHG detection (The parametric lines increase exponentially (see Chapter III) while SHG increases quadratically with the atomic density). (5) At low densities the percentage of dimers is also reduced.

Crucial measurements are those of the intensity and density dependence of I_{SHG} , but measurements of the line shape and width would also give valuable information. The following measurements are proposed:

- (1) Intensity dependence: From the photoionization model a stronger dependence than I_L^2 is to be expected at low pump intensities. Most useful would be a simultaneous measurement of photoelectron current or fluorescence from the upper state and SHG as functions of the pump intensity. This simultaneous measurement should enable us to know with certainty when the two-photon saturation sets in and test the dc-electric field model.
- (2) A precise determination of the atomic density is needed. It can be

done by absorption, fluorescence or by weighting the amount of metal settled in a collecting film during a fixed time interval. An N^2 -dependence could be reconciled with the photoionization model only if the electric field becomes independent of the atomic density. Simultaneous measurement of the photoelectron current could give additional valuable information.

- (3) Line shape and width: By the use of a Fourier limited laser, the measurement of ref. /27/ could be highly improved. We then expect to resolve the hyperfine structure and determine the SHG line width, which is probably mainly determined by Stark broadening.
- (4) Working with Rydberg states supplies an additional advantage. A measurement of the SHG conversion efficiency as a function of the principal quantum number at low densities, where phase-mismatch effects can be neglected, should also yield a better understanding of the mixing process and reflect for example the photoionization or the l-mixing cross section, as well as the variations of the effective Rabi frequency and Stark shifts with the quantum number.
- (5) Recently Prof. Y. Shen /75/ suggested an experiment with picosecond pulsed lasers. The basic idea is that if SHG is due to a dc-field, which takes a time long compared to the length of the pulse to build up, SHG should not be observed for very short pulses, that is shorter than the inverse of the plasma frequency. If instead it is due to some intrinsic atomic or atom-field interaction properties, it should be observed even for very short pulses.

APPENDIX I

DERIVATION OF EQUATIONS FOR THE RESONANT DENSITY MATRIX ELEMENTS

The electric field is given in terms of the slowly varying envelopes $\tilde{E}_\alpha(z, t)$ by Eq. (2.1). We will be first concerned with time dependences; therefore let us introduce the complex amplitude $E_\alpha(z, t)$ defined by

$$E_\alpha(z, t) = \tilde{E}_\alpha(z, t) \exp(ik_\alpha z), \quad (A1.1)$$

which is slowly varying in time but not in space.

Equations for the slowly varying amplitudes A_{lj}^{NLM} are obtained by inserting the Fourier expansion (2.7) into the density matrix equation (2.6):

$$\begin{aligned} \dot{A}_{lj}^{NLM} = & -i(\omega_{lj} + \nu_{NLM}) A_{lj}^{NLM} + \sum_{km} R_{ljkm} A_{km}^{NLM} \delta(\nu_{N'L'M'} - \nu_{NLM}) \\ & + 1/2\hbar \sum_{\alpha=1}^3 [d \cdot A^{N'L'M'}]_{lj} (F_\alpha)_{N'L'M'}^{NLM} \end{aligned} \quad (A1.2)$$

where we have introduced the function

$$(F_\alpha)_{N'L'M'}^{NLM} = E_\alpha \delta(\nu_{N'L'M'} - \nu_{NLM} - \nu_\alpha) + E_\alpha^* \delta(\nu_{N'L'M'} - \nu_{NLM} + \nu_\alpha) \quad (A1.3)$$

for convenience we have adopted Einstein's convention for summing over the repeated indexes $N'L'M'$. The sum remains everywhere a primed sum.

We first solve Eq. (A1.2) for the nonresonant amplitudes; to avoid confusion we designate nonresonant and resonant amplitudes B_{lj}^{NLM} and ρ_{lj}^{NLM} , respectively (resonant are those satisfying Eq. (2.10)). By

neglecting: (1) any coupling between nonresonant amplitudes, and (2) the coupling between resonant and nonresonant amplitudes through the signal/idler fields, the sum over α in Eq. (A1.2) can be reduced to one term, that corresponding to the pump field, i.e., $\alpha=1$, and we have

$$\dot{B}_{lj}^{NLM} = -i\Delta_{lj}^{NLM} B_{lj}^{NLM} + 1/2\hbar [d, \rho^{N'L'M'}]_{lj} (F_1)_{N'L'M'}^{NLM} \quad (A1.4)$$

where the quantities

$$\Delta_{lj}^{NLM} = \omega_{lj} + \nu_{NLM} - i\gamma_{lj}(1-\delta_{lj}) + i\Gamma_l\delta_{lj} \approx \omega_{lj} + \nu_{NLM} \quad (A1.5)$$

are essentially nonresonant detunings.

We next transform Eq. (A1.4) into its integral form

$$B_{lj}^{NLM}(t) = i/2\hbar \int_{-\infty}^t dt' \exp(-i\Delta_{lj}^{NLM}(t-t')) [d, \rho^{N'L'M'}]_{lj} (F_1)_{N'L'M'}^{NLM} \quad (A1.6)$$

and integrate by parts assuming that $\mathcal{E}_l(t)$ and all its derivatives vanish for $t \rightarrow -\infty$:

$$B_{lj}^{NLM}(t) = (i/2\hbar \Delta_{lj}^{NLM}) \sum_{s=0}^{\infty} (i/\Delta_{lj}^{NLM})^s \frac{d^s}{dt^s} \left\{ [d, \rho^{N'L'M'}]_{lj} (F_1)_{N'L'M'}^{NLM} \right\}. \quad (A1.7)$$

The infinite series in Eq. (A1.7) converges rapidly if the temporal change of the amplitude $\mathcal{E}_l(t')$ is slow compared with that of the exponential in Eq. (A1.6). From Eq. (A1.7) we can see roughly that, if $|\mathcal{E}_l| \gg |1/\Delta_{lj}^{NLM} d\mathcal{E}_l/dt|$, then it is enough to retain only the first term in the sum. We will assume that this adiabatic criterion, usually written as

$$|d\mathcal{E}_l/dt| \ll |\Delta_{lj}^{NLM} \mathcal{E}_l|. \quad (A1.8)$$

is satisfied and take

$$B_{lj}^{NLM}(t) = (1/2\hbar \Delta_{lj}^{NLM}) [d, \rho^{N'L'M'}]_{lj} (F_1)_{N'L'M'}^{NLM}. \quad (A1.9)$$

The adiabatic criterion can also be expressed in terms of the pump pulse duration τ as $\Delta_{lj}^{NLM} \gg 1/\tau$, by taking $|dE/dt| \sim |E_s/\tau|$. That this condition be fulfilled is actually our main assumption.

We now go back to Eq. (A1.2) to find the equations for the resonant amplitudes; we divide the sums into resonant and nonresonant contributions. The first sum does not couple terms with different frequencies, and therefore does not include nonresonant amplitudes. We have

$$\begin{aligned} \dot{\rho}_{lj}^{NLM} = & -i(\omega_{lj} + \nu_{NLM}) \rho_{lj}^{NLM} + \sum_{km} R_{ljkm} \rho_{km}^{N'L'M'} \delta(\nu_{N'L'M'} - \nu_{NLM}) \\ & + i/2\hbar \sum_{\alpha=2,3} [d, \rho^{N'L'M'}]_{lj} (F_\alpha)_{N'L'M'}^{NLM} \\ & + i/2\hbar [d, B^{N'L'M'}]_{lj} (F_\alpha)_{N'L'M'}^{NLM}. \end{aligned} \quad (A1.10)$$

In the sum over resonant amplitudes use has been made of the fact that no pair of resonant frequencies $\nu_{N'L'M'}$, ν_{NLM} satisfy $\nu_{N'L'M'} - \nu_{NLM} = \pm \nu_i$. They therefore differ either by one signal/idler photon or two pump photons, but not by one. A similar situation occurs in the sum over nonresonant amplitudes. From eqs. (A1.2) and (A1.9), the nonresonant frequency must simultaneously satisfy

$$\nu_{N'L'M'} = \nu_{NLM} \pm \nu_1$$

$$\nu_{N'L'M'} = \nu_{N''L''M''} \pm \nu_\alpha, \quad \alpha=1,2,3, \quad (\text{A1.11})$$

where ν_{NLM} and $\nu_{N''L''M''}$ are resonant frequencies. This is possible only for $\alpha=1$. Therefore no cross terms $\mathcal{E}_1\mathcal{E}_2$ or $\mathcal{E}_1\mathcal{E}_3$ will appear in the final equations of motion for the resonant amplitudes. They are obtained by substituting Eq. (A1.9) into Eq. (A1.10) to get

$$\begin{aligned} \dot{\rho}_{lj}^{NLM} = & -i(\omega_{lj} + \nu_{NLM}) \rho_{lj}^{NLM} + \sum_{km} R_{ljkm} \rho_{km}^{N'L'M'} \delta(\nu_{N'L'M'} - \nu_{NLM}) \\ & + i/2\kappa \sum_{\alpha=2,3} [d, \rho^{N'L'M'}]_{lj} (F_\alpha)_{N'L'M'}^{NLM} \end{aligned} \quad (\text{A1.12})$$

$$\begin{aligned} & + 1/(2\kappa) (S_{lj}^{N'L'M'} |\mathcal{E}_1|^2 \delta(\nu_{N'L'M'} - \nu_{NLM}) + \\ & + T_{lj}^{N'L'M'} (-\nu_1) \mathcal{E}_1^2 \delta(\nu_{N'L'M'} - \nu_{NLM} - 2\nu_1) \\ & + T_{lj}^{N'L'M'} (+\nu_1) \mathcal{E}_1^2 \delta(\nu_{N'L'M'} - \nu_{NLM} + 2\nu_1)) \end{aligned}$$

where $S_{lj}^{N'L'M'}$, $T_{lj}^{N'L'M'}$ are coefficients giving the two-photon Stark shifts and transition probabilities respectively, and are given by

$$S_{lj}^{N'L'M'} = \sum_k \frac{2d_{lk} [d, \rho^{N'L'M'}]_{kj} (\omega_{kj} + \nu_{N'L'M'})}{(\omega_{kj} + \nu_{N'L'M'})^2 - \nu_1^2} - \frac{2[d, \rho^{N'L'M'}]_{lk} d_{kj} (\omega_{lk} + \nu_{N'L'M'})}{(\omega_{lk} + \nu_{N'L'M'})^2 - \nu_1^2} \quad (\text{A1.13})$$

$$T_{lj}^{N'L'M'}(\mp \nu_1) = \sum_k \frac{d_{lk} [d, \rho^{N'L'M'}]_{kj}}{\omega_{kj} + \nu_{N'L'M'} \mp \nu_1} - \frac{[d, \rho^{N'L'M'}]_{lj} d_{kj}}{\omega_{lk} + \nu_{N'L'M'} \mp \nu_1}$$

The set of three-level density matrix equations in their usual form

are easily obtained from Eq. (A1.12), remembering the resonance condition given by Eq. (2.10), and the fact that only d_{1j} , d_{j2} , with $j \geq 3$ are different from zero :

$$\begin{aligned}
 \dot{\rho}_{21}^{-200} &= -(\gamma_{21} + i\Delta_1 + i\omega_{s1}I_1) \rho_{21}^{-200} + i\rho_{23}^{0-10} V_{31} - iV_{23} \rho_{31}^{00-1} + iV_{21} (\rho_{22}^{000} - \rho_{11}^{000}) \\
 \dot{\rho}_{23}^{0-10} &= -(\gamma_{23} + i\Delta_2 + i\omega_{s2}I_1) \rho_{23}^{0-10} + i\rho_{21}^{-200} V_{13} - iV_{21} \rho_{13}^{001} + iV_{23} (\rho_{22}^{000} - \rho_{33}^{000}) \\
 \dot{\rho}_{13}^{001} &= -(\gamma_{13} - i\Delta_3 - i\omega_{s3}I_1) \rho_{13}^{001} + i\rho_{12}^{200} V_{23} - iV_{12} \rho_{23}^{0-10} - iV_{13} (\rho_{33}^{000} - \rho_{11}^{000}) \\
 \dot{\rho}_{11}^{000} &= \gamma_3 \rho_{33}^{000} + (i\rho_{12}^{200} V_{21} + iV_{31} \rho_{13}^{001} + \text{c.c.}) \\
 \dot{\rho}_{22}^{000} &= -\gamma_2 \rho_{22}^{000} - (i\rho_{12}^{200} V_{21} + i\rho_{32}^{010} V_{23} + \text{c.c.}) \\
 \dot{\rho}_{33}^{000} &= -\gamma_3 \rho_{33}^{000} + \gamma_2 \rho_{22}^{000} + (i\rho_{32}^{010} V_{23} + iV_{13} \rho_{31}^{00-1} + \text{c.c.})
 \end{aligned} \tag{A1.14}$$

The following quantities have been introduced in Eq. (A1.14): the reduced interaction matrix elements $V_{23} = -d_{23} \mathcal{E}_2 / 2\hbar$,

$$V_{31} = -d_{31} \mathcal{E}_3 / 2\hbar, \quad V_{21} = k_{21} \mathcal{E}_1^2 / 2\hbar \quad \text{with} \quad k_{21} = 1/2\hbar \sum_j d_{2j} d_{j1} / (\omega_{2j} - \nu_1);$$

the dimensionless intensity $I_1 = 2|V_{21}| \sqrt{T_1 / \gamma_{21}}$ with $T_1 = 1/2 (2/\gamma_2 + 1/\gamma_3)$, and the two-photon Stark shifts,

$$\begin{aligned}
 \omega_{s1} &= -1/2\hbar \sum_j \frac{|d_{1j}|^2 \omega_{1j}}{\omega_{1j}^2 - \nu_1^2} + \frac{|d_{2j}|^2 \omega_{j2}}{\omega_{j2}^2 - \nu_1^2} (1/k_{21} \sqrt{\gamma_{21} / T_1}) \\
 \omega_{s2} &= -1/2\hbar \sum_j \frac{|d_{3j}|^2 \omega_{3j}}{\omega_{3j}^2 - \nu_1^2} + \frac{|d_{j2}|^2 \omega_{j2}}{\omega_{j2}^2 - \nu_1^2} (1/k_{21} \sqrt{\gamma_{21} / T_1}) \\
 \omega_{s3} &= \omega_{s1} - \omega_{s2}
 \end{aligned} \tag{A1.15}$$

To obtain a closed system we have assumed $\Gamma_2 = \Gamma_{23} \equiv \gamma_2$, $\Gamma_3 = \Gamma_{31} \equiv \gamma_3$, and neglected all other population relaxation rates.

In Eq. (A1.14) the V 's and ρ^{NM} 's are not slowly varying in z . To remove the rapid spatial dependence, we make the following transformation:

$$\begin{aligned}
 \rho_{jj}^{000} &= \tilde{\rho}_{jj} \quad , \quad j=1,2,3 \\
 \rho_{21}^{-200} &= \tilde{\rho}_{21} \exp(2ik_1 z) \\
 \rho_{23}^{0-10} &= \tilde{\rho}_{23} \exp(ik_2 z) \\
 \rho_{13}^{001} &= \tilde{\rho}_{13} \exp(-ik_3 z) \\
 \tilde{\rho}_{ji} &= \tilde{\rho}_{ji}^*
 \end{aligned} \tag{A1.16}$$

The transformed equations then relate only slowly varying quantities in time as well as in space :

$$\begin{aligned}
 \dot{\tilde{\rho}}_{21} &= -(\gamma_{21} + i\Delta_1 + i\omega_{s1}I_1)\tilde{\rho}_{21} + (i\tilde{\rho}_{23}\tilde{V}_{31} - i\tilde{V}_{23}\tilde{\rho}_{31})\exp(-i\Delta kz) + i\tilde{V}_{21}(\tilde{\rho}_{22} - \tilde{\rho}_{11}) \\
 \dot{\tilde{\rho}}_{23} &= -(\gamma_{23} + i\Delta_2 + i\omega_{s2}I_1)\tilde{\rho}_{23} + (i\tilde{\rho}_{21}\tilde{V}_{13} - i\tilde{V}_{21}\tilde{\rho}_{13})\exp(i\Delta kz) + i\tilde{V}_{23}(\tilde{\rho}_{22} - \tilde{\rho}_{33}) \\
 \dot{\tilde{\rho}}_{13} &= -(\gamma_{13} - i\Delta_3 - i\omega_{s3}I_1)\tilde{\rho}_{13} + (i\tilde{\rho}_{12}\tilde{V}_{23} - i\tilde{V}_{12}\tilde{\rho}_{23})\exp(-i\Delta kz) - i\tilde{V}_{13}(\tilde{\rho}_{33} - \tilde{\rho}_{11}) \\
 \dot{\tilde{\rho}}_{11} &= \gamma_3\tilde{\rho}_{33} + (i\tilde{\rho}_{12}\tilde{V}_{21} + i\tilde{V}_{31}\tilde{\rho}_{13} + \text{c.c.}) \\
 \dot{\tilde{\rho}}_{22} &= -\gamma_2\tilde{\rho}_{22} - (i\tilde{\rho}_{12}\tilde{V}_{21} + i\tilde{\rho}_{32}\tilde{V}_{23} + \text{c.c.}) \\
 \dot{\tilde{\rho}}_{33} &= -\gamma_3\tilde{\rho}_{33} + \gamma_2\tilde{\rho}_{22} + (i\tilde{\rho}_{32}\tilde{V}_{23} + i\tilde{V}_{13}\tilde{\rho}_{31} + \text{c.c.})
 \end{aligned} \tag{A1.17}$$

where $\tilde{V}_{23} = -d_{23}\tilde{\epsilon}_2/2\kappa$, $\tilde{V}_{31} = -d_{31}\tilde{\epsilon}_3/2\kappa$, $\tilde{V}_{21} = k_{21}\tilde{\epsilon}_1^2/2\kappa$, and $\tilde{V}_{ij} = \tilde{V}_{ji}^*$. The quantity $\Delta k = 2k_1 - k_2 - k_3$ in the exponential describes the phase mismatch due to the different wave velocities in the medium.

REFERENCES.

- /1/ P.A. Franken, A.E.Hill, C.W.Peters, G.Weinreich, Phys.Rev.Lett. 7 , 118, (1961).
- /2/ P.D.Maker, R.W.Terhune, M.Nisenoff, C.M.Savage, Phys.Rev.Lett. 8 , 21, (1962).
- /3/ G.H.C.New, J.F.Ward, Phys.Rev.Lett. 19 , 556 (1967).
- /4/ J.F.Ward G.H.C.New, Phys.Rev. 185 , 57 (1969).
- /5/ S.E.Harris, R.B.Miles, Appl.Phys.Lett. 19 , 385 (1971).
- /6/ R.B.Miles, S.E.Harris, IEEE J. QE-9 , 470 (1973).
- /7/ J.F.Young, G.C.Bjorklund, A.H.Kung, R.B.Miles, S.E.Harris, Phys. Rev.Lett. 27 , 1551 (1971).
- /8/ D.C.Hanna, M.A.Yuratich, D.Cotter, "Nonlinear optics of free Atoms and Molecules", Springer Verlag, Berlin, Heidelberg, NY 1979.
- /9/ Q.H.F. Vrehen and H.M.J.Hikspoors. Opt.Comm. 21 , 127 (1977).
- /10/ D.Cotter, D.C.Hanna, W.H.W.Tuttlebee and M.A.Yuratich, Opt.Comm. 22 , 190 (1977).
- /11/ J.Reif and H.Walther, Appl.Phys 15 , 361 (1978).
- /12/ J.Heinrich. Dissertation, Universitaet Düsseldorf (1983).
- /13/ J.A.Hermann and B.V.Thompson, J.Phys.B 14 , 2961 (1981).
- /14/ W.H.Louisell and P.Meystre, Phys.Rev.A 23 , 1015 (1981).
- /15/ J.R.Taylor, Opt. Commun. 18 , 504 (1976).
- /16/ A.V.Smith and J.F.Ward, IEEE J. QE-17 , 525 (1980).
- /17/ S.Dinev, Priv. Comm. 1983.
- /18/ R.S.Finn and J.W.Ward, Phys. Rev.Lett. 26 , 285 (1971);
I.J.Bigglo and J.W.Ward, Phys.Rev.A 9 , 35 (1974).
- /19/ D.P.Shelton and A.D.Buckingham, Phys.Rev. A 26 , 2787 (1982)
- /20/ D.S.Bethune, R.W.Smith and Y.R.Shen, Phys.Rev.Lett. 37 , 431 (1976) and Phys.Rev. A. 17 , 277 (1978).
- /21/ A.Flusberg, T.Mossberg, S.R.Hartmann, Phys.Rev.Lett 38 , 59 (1977);

- M.Matsuoka, H.Nakatsuka, H.Uchiki, M.Mitsunaga, Phys.Rev.Lett. 38 894 (1977); H.Uchiki, H.Nakatsuka, M.Matsuoka, Opt.Comm. 30 , 345 (1979).
- /22/ T.Mossberg, A.Flusberg, and S.R.Hartmann, Opt.Comm. 25 , 121 (1978).
- /23/ K.Miyasaki, T.Sato, H.Kashiwagi, Phys.Rev. A 23 , 1358 (1981).
- /24/ R.R.Freeman, L.E.Bjorkholm, R.L.Panock, in Laser Spectroscopy V, ed. by A.R.W.Mckellar, T.Oka, and B.P.Stoicheff, Springer Verlag 1981, p.453.
- /25/ W.Jamroz, P.E.LaRocque, and B.P.Stoicheff, Opt.Lett. 7 , 148 (1982).
- /26/ J.Okada, Y.Fukuda, M.Matsuoka, J.Phys.Soc.Japan 50 , 1301 (1981).
- /27/ S.Dinev, A.Guzman de Garcia, P.Meystre, R.Salomaa, and H.Walther. Laser Spectroscopy VI, ed. by H.P.Weber and W.Luthy. Springer Verlag 1983, p.183.
- /28/ D.S.Bethune, Opt.Lett. 6 , 287 (1981).
- /29/ D.S.Bethune, Phys.Rev. A 23 , 3139 (1981).
- /30/ J.Heinrich and W.Behmenburg, Appl.Phys. 23 , 333 (1980).
- /31/ M.Ducloy, Y.Leite and M.S.Feld, Phys.Rev. A 17 , 623 (1978).
- /32/ L.Allen, J.H.Eberly, "Optical resonance and two-level atoms". Wiley NY 1975, and references therein.
- /33/ R.P.Feynman, F.L. Vernon Jr. and R.W.Hellwarth, J.Appl.Phys. 28 , 49 (1957).
- /34/ A.Guzman de Garcia, P.Meystre and M.Sargent III, Opt.Comm. 43 , 364 (1982).
- /35/ H.G. von Garssen, Private Comm. 1983.
- /36/ R.G.Brewer and E.L.Hahn, Phys.Rev.A 11 , 1641 (1975).
- /37/ T.Haensch and P.Toschek, Z.Phys. 236 , 213 (1970).
- /38/ I.M.Beterov and V.P.Chebotaev in "Progress in Quantum Electronics 3", ed by J.H.Sanders and S.Stenholm (Pergamon, Oxford, 1974).

- /39/ Y.R. Shen, Rev.Mod.Phys. 48 , 1 (1976).
- /40/ R.Salomaa and S.Stenholm, J.Phys. B 9 , 1221 (1976).
- /41/ P.R.Berman and R.Salomaa, Phys.Rev. A 25 , 2667 (1982)
- /42/ E.Kyrola and R.Salomaa, Phys.Rev. A 23 , 1874 (1981) and
Appl.Phys. 20 ,339 (1979).
- /43/ M.Goppert-Mayer, Ann.Phys. (Leipz.) 9 , 273 (1931).
- /44/ M.Takatsuji, Phys.Rev. A 11 , 619 (1975).
- /45/ D.Grischkowsky, M.M.Loy, and P.F.Liao, Phys.Rev.A 12 , 251 (1975).
- /46/ D.Grischkowsky, R.G.Brewer, Phys.Rev.A 15 , 1789 (1977).
- /47/ See for ex. M.Sargent III, M.O.Scully, and W.E.Lamb, "Laser
Physics", ed. Addison-Wesley, Mass. 1974.
- /48/ Autler S.H., Townes C.H., Phys.Rev. 100 ,707 (1955);
Javan A., Phys.Rev. 107 ,1579 (1957);
Kontorovich V.M., Prokhorov A.M.,Sov.Phys.JETP 6 , 1100 (1958);
Brunner W.,Paul H.,Richter G., Steudel H., Ann. Physik 21 , 187
(1968); Steudel H., Ann. Physik 22 ,113 (1969).
- /49/ J. Heppner, C.D.Weiss, U.Huebner, G.Schirm, J.of QE-16 , 392
(1980).
- /50/ Yu.A.Il'inskii and V.D.Taranukhin, Sov.J.Quant.Electron. 4 , 997
(1975).
- /51/ D.Y.Kim and P.D.Coleman, IEEE J. QE-16 ,300 (1980).
- /52/ S.J.Petuchowski and J.D.Oberstar, T.A. DeTemple, Phys.Rev.A 20 ,
529 (1979).
- /53/ B.Hocker and C.L.Tang, Phys.Rev. 184 , 356 (1969);
R.G.Brewer and R.L.Shoemaker Phys.Rev.Lett. 27 , 631 (1971).
- /54/ A.Schenzle and R.G.Brewer, Phys.Rev. A 14 , 1756 (1976);
Phys.Rep. 43 , 455 (1978).
- /55/ R.L.Shoemaker and R.G.Brewer, Phys.Rev.Lett. 28 , 1430 (1972).
- /56/ R.G.Brewer and R.L.Shoemaker, Phys.Rev. A 6 , 2001 (1972)
- /57/ N.A.Kurnit, I.D.Abella and S.R.Hartmann, Phys.Rev.Lett. 13 , 567

- (1964); C.K.N.Patel and R.E.Slusher, Phys.Rev.Lett. 20 , 1089 (1968).
- /58/ T.W.Mossberg, R.Kachru, S.R.Hartmann and A.M.Flusberg, Phys.Rev.A 20 , 1976 (1979).
- /59/ N.Tan-no, K.Yokoto and H.Inaba, Phys.Rev.Lett. 29 , 1211 (1972).
- /60/ M.S.Feld in 'Frontiers in Laser Spectroscopy' ed. by R.Balian, S.Haroche and S.Liberman. (North-Holland, Amsterdam).
- /61/ F.S. Acton, 'Numerical methods that work', Harper & Row publishers. N.Y., Evanston and London (1970).
- /62/ P.Meystre, M.D.Scully, and H.Walther, Optics Com. 33 , 153 (1980).
- /63/ A.Gallagher, T.Holstein, Phys.Rev.A 16 , 2413 (1977); S.Geltman, J.Phys.B 10 , 3057 (1974).
- /64/ N.Allard and J.Kielkopf, Rev.Mod.Phys. 54 , 1103 (1982), and references therein.
- /65/ J.Pascale, J. of Chem.Phys. 67 , 204 (1977).
- /66/ T.F.Gallagher and W.E.Cooke, Phys.Rev.A 19 , 2161 (1979).
- /67/ A.Omont, J. de Phys. 38 , 1343 (1977).
- /68/ J.Derouard and M.Lombardi, J.Phys.B 11 , 3875 (1978); A.P.Hickman, Phys.Rev.A 18 , 1339 (1978); *ibid.* 19 , 994 (1979); *ibid.* 23 , 87 (1981). M.Chapelet, J.Boulmer, J-G.Gauthier and J-F.Delpech, J.Phys.B 15 , 3455 (1982); J.I.Gersten, Phys.Rev.A 14 , 1354 (1976).
- /69/ M.Hugon, F.Gounand, P.R.Fournier, and J.Berlande, J.Phys.B 12 , 2707 (1979).
- /70/ M.Hugon, F.Gounand, P.R.Fournier, J.Phys.B 14 , L109 (1980); M.Hugon, F.Gounand, P.R.Fournier, and J.Berlande, J.Phys.B 16 , 2531 (1983).
- /71/ J.M.Raimond, G.Vitrant and S.Haroche, J.Phys.B 14 , L655 (1981).
- /72/ D.R.Bates and A.Damgaard, Phil.Trans.Roy.Soc.London A242 , 101 (1949).

- /73/ B.Quarder, Ann.Physik 74 , 255 (1924).
- /74/ J.A.Armstrong, N.Bloembergen, J.Ducuing, and P.S.Perehan, Phys.Rev. 127 , 1918 (1962).
- /75/ R.Shen, Priv. Comm. (1984)
- /76/ C.C.Wang and I.V.James, J.-F.Xia, Laser Spectroscopy VI, ed. by H.P.Weber and W.Luthy. Springer Verlag 1983, p.356.
- /77/ D.Rogovin and A.Elci, and P.Avizonis, Proc.Int.Conf.Lasers'80, New Orleans, Louisiana USA.
- /78/ P.W.Anderson, Phys.Rev. 76 , 647 (1949).

ACKNOWLEDGEMENTS

I gratefully acknowledge the aid and encouragement of Dr. habil. P. Meystre and Professor Dr. H. Walther, without whose invaluable support, none of this work could have been accomplished. I am also deeply indebted to Professor R. Salomaa; his sustained interest and efforts on my behalf were a source of great inspiration. I would also like to thank these gentlemen as well as Professor Dr. H. Wolter for many provoking discussions and a critical review of this work.

My gratitude to Professor M. Sargent III, with whom I obtained the first analytical results, and to the experimentalists Professor Dr. W. Behmenburg and Dr. J. Heinrich at the Universität Düsseldorf, Dr. S. Dinev at the Max Planck Institut für Quantenoptik for their promptness and collaboration in discussing their experiments.

I also gained a lot from the encouragement of friends in "The Theoretical Container" of the MPQ where this work was carried out, especially to Drs. J. Gallas, P. Dobiasch and D. P. O'Brien, who also read and commented upon part of this work. He is responsible for the "Irish Accent" at some points.

My thanks to C. S. Cha for his advise in solving some computing problems and to Fr. Pfister for her efficient drawings.

This work was made possible through much appreciated financial support from the Deutscher Akademischer Austauschdienst, and the Universidad Nacional de Colombia (Bogotá, Colombia).

LEBENS LAUF

NAME, VORNAME : Guzmán de García Angela María.

GEBURTSNAME : Guzmán.

GEBURTSDATUM : 4. 12. 1954.

GEBURTSORT : Bogotá. Kolumbien.

FAMILIENSTAND : Verheiratet.

BERECHTIGUNG ZUM HOCHSTUDIUM

GYMNASIUM : Colegio Santa Clara

SCHULORT : Bogotá. Kolumbien.

ZEITRAUM: Feb. 1964 - Nov. 1969.

ART DER BERECHTIGUNG : Bachiller.

DATUM : 24. 11. 1969.

HOCHSTUDIUM

HOCHSCHULE : Universidad Nacional de Colombia.

ORT : Bogotá. Kolumbien.

STUDIENFACH : Physik.

ZEITRAUM : Feb. 1970 - März 1975.

DIPLOM : Físico.

DATUM : 14. 3. 1975.

POSTGRADUIERTENSTUDIUM

HOCHSCHULE : Universidad Nacional de Colombia.

ORT : Bogotá. Kolumbien.

STUDIENFACH : Physik.

ZEITRAUM : Mai 1975 - März 1981

DIPLOMARBEIT : "Aplicación del método de parámetro de impacto al cálculo de secciones eficaces en el sistema protón-hidrógeno."

BETREUER : Dr. rer. nat. D. Campos.

DIPLOM : Magister Scientiae mit Schwerpunkt in Physik.

DATUM : 27. 2. 1981.

VERÖFFENTLICHUNGEN

'Total cross sections for resonance charge transfers between $H(1s)$ and H^+ calculated by using a two-state orthonormal basis with static variable charge'. D.Campos, C.Ramírez and A. de García, J.Phys.B **16** , 853 (1983).

'Probe gain in a Two-Photon pumped Three-level System'. A.Guzmán de García, P.Meystre and M.Sargent III, Opt.Comm. **43** , 364 (1982).

'Subnatural Spectroscopy'. A.Guzmán, P.Meystre and M.D.Scully. ADVANCES IN LASER SPECTROSCOPY, ed. by F.T.Arechí, F.Strumia, and H. Walther. Plenum Publishing Corporation 1983, p.465.

'Role of collisions in second harmonic generation in alkali vapors'. S.Dinev, A.Guzmán de García, P.Meystre, R.Salomaa, and H. Walther. LASER SPECTROSCOPY VI, ed. by H.P.Weber and W.Luthy. Springer Verlag 1983, p.183.

